THE PHYSICS OF NUCLEAR REACTORS

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Foreword

THE papers published in this supplement, with the discussion on them, comprise the proceedings of a Conference arranged by The Institute of Physics in collaboration with the British Nuclear Energy Conference and held in London from 3 to 6 July, 1956.

The purpose of the Conference was to keep members of the Institute and its sister Institutions abreast of scientific developments in nuclear reactors and to call their attention to the problems needing solution. The Conference was particularly intended for industrial and government physicists, working or likely to work directly or indirectly with reactors, and for lecturers and other teachers. Close on five hundred persons attended the Conference, including many from oversea.

I commend this supplement as a useful record of the present state of our knowledge and of the gaps which need to be filled.

J. D. Cockcroft

President, The Institute of Physics

The Physics of Nuclear Reactors

Errata

p. iv For *Peace Time Uses of Atomic Energy* read *Peaceful Uses of Atomic Energy*.

p. S 48 The ordinate of Fig. 7 should read g/mm^2 *not* kg/mm^2.

CONTENTS

Session 1. Opening address

Page

Scientific problems in the development of nuclear power. By SIR JOHN COCKCROFT, K.C.B., C.B.E., D.Sc., F.Inst.P., F.R.S., Atomic Energy Research Establishment, Harwell — S 1

Session 2. Lecture

The physics of advanced reactors. By J. R. DIETRICH, Ph.D., Argonne National Laboratory, U.S.A. — S 9

DISCUSSION — S 24

Session 3. Lectures

Experimental reactor physics. By P. W. MUMMERY, M.A., Atomic Energy Research Establishment, Harwell — S 27

The zero-energy fast breeder reactor, ZEPHYR. By L. R. SHEPHERD, B.Sc., Ph.D., A.Inst.P., Atomic Energy Research Establishment, Harwell — S 32

DISCUSSION — S 41

Session 4. Evening lecture

Recent developments in nuclear theory. By Prof. R. E. PEIERLS, C.B.E., D.Sc., F.R.S., University of Birmingham (Headings only) — S 42

Session 5. Lecture

The effects of irradiation on the physical properties of solids. By A. H. COTTRELL, B.Sc., Ph.D., F.R.S., Atomic Energy Research Establishment, Harwell — S 43

DISCUSSION — S 49

Session 6. Discussion on "Ancillary problems of reactors"

OPENING PAPERS

Shielding for reactors. By G. C. LAURENCE, M.B.E., Ph.D., F.R.S.C., Atomic Energy of Canada Ltd., Chalk River, Ontario — S 54

The control of reactors. By A. J. Salmon, B.Sc., Ph.D., A.M.I.E.E., A.Inst.P., Associated Electrical Industries Ltd., Aldermaston — S 59

The instrumentation of reactors. By W. MACRAE, M.Sc., A.M.I.E.E., C. A. Parsons and Co. Ltd., Newcastle upon Tyne — S 71

DISCUSSION — S 82

Session 7. Evening lectures

Page

The role of chemistry in a nuclear energy project. By R. SPENCE, C.B., D.Sc., F.R.I.C., Atomic Energy Research Establishment, Harwell S 88

The role of metallurgy in a nuclear energy project. By L. ROTHERHAM, M.Sc., F.I.M., F.Inst.P., Industrial Group Headquarters, U.K. Atomic Energy Authority, Risley S 91

Session 8. Discussion on "Research reactors and their uses"

OPENING PAPERS

The Canadian research reactors and their uses. By W. B. LEWIS, C.B.E., Ph.D., F.R.S.C., F.R.S., Atomic Energy of Canada Ltd., Chalk River, Ontario S 96

The United Kingdom research reactors and their uses. By F. W. FENNING, B.A., Atomic Energy Research Establishment, Harwell S 100

DISCUSSION S 103

EXHIBITS AND FILMS S 107

SUBJECT INDEX S 108

NAME INDEX S 111

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United Nations Organization, publishers of the *Proceedings of the International Conference on the Peace Time Uses of Atomic Energy* held in Geneva in 1955:

Figs. 4, 5, 6 (S 4); 9, 10, 11 (S 6); 13 (S 8); 1 (S 10); 2 (S 11); 3 (S 13); 9, 10 (S 19); 14 (S 21); 1 (S 28); 4, 5 (S 30); 1 (S 33); 2 (S 34); 3, 4 (S 38); 1 (S 44); 2, 3 (S 45); 5 (S 46); 6 (S 47).

Atomic Energy Research Establishment, Harwell:

Figs. 1, 2 (S 2); 7, 8 (S 5); 12 (S 7); 5 (S 38); 1, 2 (S 72); 3 (S 73).

Atomic Energy of Canada Ltd., Chalk River, Ontario:

Figs. 1 (S 54); 2, 3 (S 55); 4, 5 (S 56); 1 (S 97); 2 (S 98).

Progress in Nuclear Energy:

Fig. 6 (S 40).

Physical Review:

Fig. 4 (S 46).

Journal of Polymer Science:

Figs. 1, 2, 3 (S 49); 4, 5 (S 50).

Nature:

Fig. 6 (S 50).

Proceedings of The Royal Society:

Fig. 7 (S 51).

Proceedings of The Institution of Electrical Engineers:

Fig. 1 (S 83).

INDEX TO THE PHYSICS OF NUCLEAR REACTORS

SUBJECT INDEX

Absorbers, parasitic S12
Absorption cross-sections S13
Aircraft, applications of reactors to S87
Assemblies
  , critical high-power S31
  , critical low-power S31
  , near-critical S31
  , sub-critical S31
Australia, plans for nuclear power in S26, S104

Breeding S24, S38

Canada, plans for nuclear power in S24
Canadian research reactors and their uses S96
Canning S93
Chemical
  changes due to irradiation S52
  problems S6
  processing of irradiated material S90
Chemistry
  in a nuclear energy project, the role of S88
  of nuclear reactors S89
Collisions, displacement S44
Computer, analogue S105
Control
  , methods of S64
  of reactors S59, S99
Controls, "signal arm" S104, S106
Conversion ratio of natural uranium reactors S12
Coolant flow S77
Coolants, radioactive isotopes produced by S58
Core structure S1
Counter, BF_3 proportional S82
Counters, pulse, measuring channels using S75
Costs S26, S104, S105
Criticality
  condition, the S27
  experiments S29
  of natural-uranium reactors S10

Danger coefficient measurements S18
Delayed neutrons S36, S40, S79, S82
Detectors S71, S82
Deuteron irradiation S53
Diffusion
  and slowing down of neutrons S29
  coefficient, changes in, due to irradiation S46
Dimensional changes due to irradiation S47
Displacement collisions S44
Disposal and utilization of radioactive wastes S90

Education and training of staff S103, S105, S106
Electric strength of polythene, effect of irradiation on S50
Electrical properties for studying radiation damage S45
Electrons, excitation of, by radiation S43
Emulsions, irradiation of S41
Energy
  distributions in ZEPHYR S37
  transfer due to irradiation S52
Enrichment
  , examples of the use of S14
  , increase of conversion ratio by S13
  , limitations on S13
  , use of, to allow higher absorption cross-sections S13
Exhibits S107

Faults in fuel containers, detection of S79
Films, exhibited S107
Fission products
  , distribution of S25
  , gaseous, detection of S80
Flow, coolant S77
Flux
  distributions in ZEPHYR S37
  measuring instruments S71
  scanning S75
France, plans for nuclear power in S105
Fuel
  containers, detection of faults in S79
  development S93
  elements and special reactor materials S89
  elements, sheathing and stability of S97
  , processing of S25

Heat
  generation, rate of S28
  output S78
Hydrocarbons, effect of irradiation on S49, S51

Instrumentation of reactors, the S71, S80
Ionization chambers, measuring circuits using S73
Irradiated material, chemical processing of S90
Irradiation
  , the effects of, on the physical properties of emulsions S41, S43, S47, S49, S51, S53
Isotopes
  , radioactive, produced by coolants S58
  , decay activity of S8

Kinetic equations of reactors S82, S84, S87

Lattices S11, S30, S99

Materials
  , raw S89
  , special reactor, fuel elements and S89
Measurements, physical S78

Measuring
channels using pulse counters S75
circuits using ionization chambers S73
Metallurgy in a nuclear energy project, the role of S91
Metals, changes in the mechanical properties of, due to irradiation S47, S51, S53
Moderators, comparison of properties of S10

Neutron
balance S2
economy-lattice experiments S99
energy spectra, the measurement of, in ZEPHYR S35
flux S56
fluxes, the measurement of, on ZEPHYR S35
leakage S29
scattering S45
sources S63
spectrum S28
Neutrons, delayed S36, S40, S60, S79, S82
Nuclear energy project
, the role of chemistry in a S88
, the role of metallurgy in a S91

Perturbation measurements, reactor S29
Perturbations in ZEPHYR S36, S39
Physical measurements S78
Physics
, experimental reactor S27
of advanced reactors, the S9
, reactor, problems S1
, solid state, problems of S5
Phosphors produced by irradiation S52
Plans for nuclear power in
Australia S26, S104
Canada S24
France S105
Soviet Union S104
U.K. S1
Poisoning
, samarium, effect of, on reactivity during operation S66
, xenon, effect of, on reactivity as a function of operating time of reactor S65
Power
measurements S71
, nuclear, scientific problems in the development of S1
Problems
, chemical S6
of solid state physics S5
, reactor physics S1
, scientific, in the development of nuclear power S1
Processing, chemical, of irradiated material S90
Pulse
counters, measuring channels using S75
some experiments S29

Radiation
damage S43, S49, S105
, excitation of electrons by S43
Radioactive wastes, disposal and utilization of S90
Raw materials S89
Reaction rates S28, S35
Reactivity
changes S2, S31, S65
measurements on ZEPHYR S36
rates S82, S87
temperature coefficients S67
Reactor
assemblies, use of S31
control S99
design S87
dynamics S27

Reactor—*continued*
kinetics S60
materials, fuel elements and special S89
materials, radiation damage in S43
physics, problems S1
physics, experimental S27
, power, essential instrumentation of a S80
power output after shut-down S64
, zero-energy fast breeder S32
Reactor
AQUILON S105
BEPO S6, S83, S100, S101
BORAX S27, S87
CP5 S64, S105
DIDO S60, S64, S83, S100, S102, S105
DIMPLE S64, S100, S102
EBR S64
EBR-I S17
EBR-II S19
EBWR S14, S64
GLEEP S100
HRE S64
LIDO S83, S100, S102, S104
MERLIN S60, S64, S104
MTR S64
NPD S24
NRU S96
NRV S98
NRX S56, S64, S96
PBR S19
PIPPA S2, S26, S60, S64
PLUTO S100, S102
SGR S14
SPERT S87
ZEPHYR S4, S17, S32, S35, S37, S60, S64, S83, S100, S103
ZEUS S4, S38, S64, S83, S100, S103
ZETR S64
ZOE S105
ZPR-III S18, S20
Reactors
, aircraft applications of S87
, boiling S16, S104
, D_2O S12
, fast S17
, fast, safety of S21
, graphite S12
, heterogeneous S83
, H_2O-moderated S15
in the Soviet Union S104
, natural-uranium criticality of S10
, nuclear, chemistry of S89
, shielding for S54
, spherical S19
, swimming-pool S104, S105
, the Canadian research, and their uses S96
, the control of S59
, the instrumentation of S71
, the physics of advanced S9
, theoretical (^{239}Pu) S83
, thermal S25, S52
, the United Kingdom research, and their uses S100
, zero-energy type S104

Safety S68, S78
Scientists, numbers employed S8
Shielding S54, S82
"Signal arm" controls S104, S106
Solid state physics, problems of S5
Solids, the effects of irradiation on the physical properties of S43
Soviet Union, plans for nuclear power in S104
Spectrometer ^3He S41
Stored energy S45

Technologists, numbers employed S8
Temperature
  coefficient feedback paths S84
  effect of irradiation S52
  measurements S76
Terminology of radiation S52
Thermal
  columns S105
  conductivity, effect of radiation on S46
Thermocouples, use of S82, S87
Thorium-^{233}U fuel S17
Training of staff, *see* Education
United Kingdom
  , plans for nuclear power in S1
  research reactors and their uses, the S100
Uranium
  , energy yield from S24
  metal, effect of irradiation on S53

Wastes, radioactive, disposal and utilization of S90
Wigner effects S6, S105

X-ray effects of irradiation S47

NAME INDEX TO THE PHYSICS OF NUCLEAR REACTORS

Absalom *see* Rose, H.
Abson, W., and Wade, F. S81
Abson, W. *see* Cox, R. J.
Alexander, P. *see* Bacq, Z. M.
Andrews, D. G. S87
Antonov, A. V. S32
Arms, H. S. S52
Avery, R. *see* Okrent, D.
Aves, R., Barnes, D., and Mackenzie, R. B. S41
Aves *see* Batchelor

Bacq, Z. M., and Alexander, P. S48
Baker, A. R., and McVicar, D. D. S41
Ballinger, H. A. *see* Jaques, T. A.
Barnes, A. H., Koch, L. J., Monson, H. O., and Smith, F. A. S23
Barnes, D. *see* Aves, R.
Barnes, R. S., and Hancock, H. H. S49
Batchelor, Aves, and Skyrme S41
Beall, S. E., and Swartout, J. A. S70
Bell, Miller, and Robson S59
Bentzen, F. L. *see* Nyer, W. E.
Berman, R., Foster, E. L., and Rosenberg, H. M. S48
Bernstien, S., Preston, W. M., Wolfe, G., and Slattery, R. E. S69
Berzelius, J. S48
Birks, J. B. S52
Birks, J. B., and Black, F. A. S53
Bisset, I. J. W. S104
Black, F. A. S53
Black, F. A. *see* Birks, J. B.
Blatt, F. J. S48
Blewitt, T. H., Coltman, R. R., Noggle, T. S., and Holmes, D. K. S48
Blewitt, T. H. *see* Thompson, D. O.
Booth, A. H. S100
Booth, A. H. *see* Hurst, D. G.
Bopp, C. D. *see* Sisman, O.
Branyan, C. E., Brunson, G. S., Cerutti, B. C., Feldes, J. G., Kato, W. Y., Kirn, F. S., Long, J. K., Porter, R. A., Rice, R. E., Okrent, D., and Thalgott, F. W. S23
Briggs, R. B., and Swartout, J. A. S70
Bright, G. O. *see* Nyer, W. E.
Brittan, R. O. S23
Broser, I., and Kallmann, H. S53
Bruch, C. A., McHugh, W. E., and Hockenbury, R. W. S49
Brunson, G. S. *see* Branyan, C. E.
Bussac, J. S105

Cahn, A. *see* Hughes, D. J.
Campbell, C. G., and Lockey, D. J. S32
Campbell, E. C. *see* Soodak, H.
Carter, J. C. *see* Spinrad, B. I.
Cerutti, B. C., Okrent, D., Rice, R. E., Thalgott, F. W., and Lichtenberger, H. V. S23
Cerutti, B. C. *see* Branyan, C. E.
Charlesby, A. S51
Charlesby, A., and Hancock, N. H. S48, S53
Charlesby, A. *see* Ross, M.
Cleland, J. W., Crawford, J. H., Lark-Horowitz, K., Pigg, J. C., and Young, F. W. S48
Cockcroft, Sir John S1
Codd, J., Shepherd, L. R., and Tait, J. H. S41
Cohen, E. R. S23
Cole, T. E. *see* Weinberg, A. M.
Coltman, R. R. *see* Blewitt, T. H.
Cooke-Yarborough, E. H., and Pulsford, E. W. S81
Cooper, H. G., Koehler, J. S., and Marx, J. W. S48
Cottrell, A. H. S5, S43, S53
Cottrell, A. H. *see* Roberts, A. C.
Cox, R. J., and Walker, J. S81, S87
Cox, R. J., Gillespie, A. B., and Abson, W. S81
Crawford, J. H., and Wittels, M. C. S48
Crawford, J. W. *see* Cleland, J. W.
Curry, M. U. *see* Holmes, J. E. R.

Dabbs, J. *see* Hughes, D. J.
Diamond, J. S103
Dienes, G. J. S48
Dietrich, J. R. S9, S26, S32, S105
Dietrich, J. R., Lichtenberger, H. V., and Zinn, W. H. S70
Downes, K. *see* Kouts, H.
Dugdale, R. A., and Green, A. S48
Dunworth, J. V. S9, S23, S25, S69

Edlund *see* Glasstone
Eggler, C. *see* Spinrad, B. I.
Ergen, W. K. *see* Weinberg, A. M.

Faris, F. E. S49
Feldes, J. G. *see* Branyan, C. E.
Fenning, F. W. S3, S4, S100, S103, S106
Fenning and Syrett S3, S4
Feshbach, H., Goertzel, G., and Yamauchi, H. S23
Forbes, S. G. *see* Nyer, W. E.
Foster, E. L. *see* Berman, R.
Franz, J. P. S87
Frisch, O. R. S41
Fuchs, K. S41

Garton, C. G. *see* Stark
Gast, P. F. S23
Geoghegan, G. R. H. S25
Gillespie, A. B. S70, S81
Gillespie, A. B. *see* Cox, R. J.
Glasstone, S. S69
Glasstone, S., and Edlund, M. C., S32, S69
Glen, J. W. S48
Glueckauf S8
Goertzel, G. *see* Feshbach, H.
Goldstein, H., and Wilkins, Jr., J. W. S59
Gorman, R. L. *see* Overhauser, A. W.
Greebler, P. S69
Green, A. *see* Dugdale, R. A.

Hall, D. *see* Hughes, D. J.
Hancock, N. H. *see* Charlesby, A.
Hanford S8
Hanna, G. C. *see* Hurst, D. G.
Harrer, J. M., Jameson, A. S., and West, J.M. S23, S70
Harrison, R. D. S87
Henderson, W. J., and Whittier, A. C. S59
Hockenbury, R. W. *see* Bruch, C. A.
Holmes S4
Holmes, D. K. *see* Blewitt, T. H.
Holmes, D. K. *see* Thompson, D. O.
Holmes, J. E. R., Curry, M. U., and Waldegrave, M. S41
Holmes, J. E. R., McVicar, D. D., Rose, H., Shepherd, L. R., Smith R. D., and Smith A. M. S41
Holmes, J. E. R., McVicar, D. D., Rose, H., Smith, R. D., and Shepherd, L. R. S23, S40, S69
Horton, C. C. S82
Hughes, D. J., Dabbs, J., Cahn, A., and Hall, D. S69
Humbach, W. S53
Hummel, H. H. *see* Okrent, D.
Hurst, D. G. S100
Hurst, D. G., Booth, A. H., Lounsbury, M., and Hanna, G. C. S32
Hurwitz, H. S69

Iskenderian, H. P., Link, L. E., Treshow, M., and West, J. M. S23

Jaques, T. A., Ballinger, H. A., and Wade, F. S81
Jameson, A. S. *see* Harrer, J. M.
Jongenburger, P. S48

Kallmann, H. *see* Broser, I.
Kato, W. Y. *see* Branyan, C. E.
Kato, W. Y. *see* Lichtenberger, H. V.
Kealy, W. A. *see* Medlock, R. S.
Keepin, G. R. S69
Kinchin, G. H., and Pease, R. S. S48
Kirn, F. S. *see* Branyan, C. E.
Kittel, J. H. *see* Paine, S. H.
Koch, L. J. *see* Barnes, A. H.
Koehler, J. S. *see* Seitz, F.
Kouts, H., Price, G., Downes, K., Sher, R., and Walsh, V. S23
Kowarski, L. S105

Lark-Horowitz, K. *see* Cleland, J. W.
Laurence, G. C. S24, S26, S54, S105
Leachman, R. B. S23
Levenson, M. S23
Lewis, W. B. S23, S96
Lichtenberger, H. V., Thalgott, F. W., Kato, W. Y., and Novick, M. S23, S70
Lichtenberger, H. V., Thalgott, F. W., and Novick, M. S41
Lichtenberger, H. V. *see* Cerutti, B. C.
Lichtenberger, H. V. *see* Dietrich, J. R.
Link, L. E. *see* Iskenderian, P. H.
Lipkin, H. J. S87
Little, D., and Schultz, M. A. S83
Littler, D. J. S69
Littler, D. J., and Raffle, J. F. S69
Lloyd, J. M., and Mills, J. E. C. S32
Lockett, G. E. S70
Lockey, D. J. *see* Campbell, C. G.
Long, J. K. *see* Branyan, C. E.
Lounsbury, M. *see* Hurst, D. G.

Mackenzie, R. B. *see* Aves, R.
Macklin, R. L., and Pomerance, H. S. S23
Macrae, W. S71, S87
Makin, M. J. S49
Mann, M. M. *see* Weinberg, A. M.
Marley, W. G., and Fry, T. M. S70
Marsden, E. S53
McEnhill, J. J. S103
McHugh, W. E. *see* Bruch, C. A.
McVicar, D. D. *see* Baker, A. R.
McVicar, D. D. *see* Holmes, J. E. R.
McVicar, D. D. *see* Smith, R. D.
Medlock, R. S., and Kealy, W. A. S81
Meyer, R. A. S48
Miller *see* Bell
Mills, J. E. C. *see* Lloyd, J. M.
Monson, H. O. *see* Barnes, A. H.
Moore, R. V. S70
Mummery, P. W. S25, S27

Noggle, T. S. *see* Blewitt, T. H.
Novick, M. *see* Lichtenberger, H. V.
Nyer, W. E., Forbes, S. G., Bentzen, F. L., Bright, G. O., Schroeder, F., and Wilson, T. R. S87

Okrent, D., Avery, R., and Hummel, H. H. S23, S41
Okrent, D. *see* Branyan, C. E.
Okrent, D. *see* Cerutti, B. C.
Overhauser, A. W. S48
Overhauser, A. W., and Gorman, R. L. S48

Paine, S. H., and Kittel, J. H. S49
Paxton, H. C. S69
Pease, R. S. *see* Kinchin, G. H.
Peierls, R. E. S42
Perkins, J. E. B. S53
Pigg, J. C. *see* Cleland, J. W.
Pomerance, H. S. *see* Macklin, R. L.
Poole, M. J. S32
Porter, R. A. *see* Branyan, C. E.
Preston, W. M. *see* Bernstien, S.
Price, G. *see* Kouts, H.
Pugh, S. F. S49
Pulsford, E. W. *see* Cooke-Yarborough, E. H.

Raffle, D. J. *see* Littler, D. J.
Rice, R. E. *see* Branyan, C. E.
Rice, R. E. *see* Cerutti, B. C.
Roberts, A. C., and Cottrell, A. H. S49
Robson *see* Bell
Rose, H. S41
Rose, H., and Absalom S41
Rose, H. *see* Holmes, J. E. R.
Rose, H. *see* Smith, R. D.
Rosenberg, H. M. *see* Berman, R.
Ross, M., and Charlesby, A. S48
Rotherham, L. S91

Salmon, A. J. S59, S81, S87, S105
Schroeder, F. *see* Nyer, W. E.
Schultz, M. A. S69
Schultz, M. A. *see* Little, D.
Seitz, F. S48
Seitz, F., and Koehler, J. S. S48
Shaw, M. *see* Simpson, J. W.
Shepherd, L. R. S32, S41, S103
Shepherd, L. R. *see* Codd, J.
Shepherd, L. R. *see* Holmes, J. E. R.
Sher, R. *see* Kouts, H.
Simpson, J. W., and Shaw, M. S70
Simpson, W. S49
Sisman, O., and Bopp, C. D. S48
Skinner, H. W. B. S26
Skvortsov, S. A. S105
Skyrme *see* Batchelor.
Slattery, R. E. *see* Bernstien, S.
Smith, A. M. *see* Holmes, J. E. R.
Smith, F. A. *see* Barnes, A. H.
Smith, R. D., McVicar, D. D., Thorne, E. A., and Rose, H. S41
Smith, R. D. *see* Holmes, J. E. R.
Smyth, H. D. S32
Soodak, H., and Campbell, E. C. S87
Spaepen, J. S52
Spence, R. S6, S88
Spinrad, B. I., Carter, J. C., and Eggler, C. S23, S100
Stark, and Garton, C. G. S53
Starr, C. S23, S69
Swartout, J. A. *see* Beall, S. E.
Swartout, J. A. *see* Briggs, J. A.
Syrett *see* Fenning

Tait, J. H. *see* Codd, J.
Taylor, B. T. S32
Taylor, D. S82
Thalgott, F. W. *see* Branyan, C. E.
Thalgott, F. W. *see* Cerutti, B. C.
Thalgott, F. W. *see* Lichtenberger, H. V.
Thomas, K. S53
Thompson, D. O., Holmes, D. K., and Blewitt, T. H. S49
Thorne, E. A. *see* Smith, R. D.
Treshow, M. *see* Iskenderian, H. P.

Untermyer, S. S23

Wade, F. *see* Abson, W.
Wade, F. *see* Jaques, T. A.
Wakelin, R. J. S87
Walker, J. *see* Cox, R. J.
Walsh, V. *see* Kouts, H.
Watson-Munro, C. N. S26
Weinberg, A. M., Cole, T. E., and Mann, M. M. S70
Weinberg, A. M., and Ergen, W. K. S87
West, J. M. *see* Harrer, J. M.
West, J. M. *see* Iskenderian, H. P.
Whittier, A. C. *see* Henderson, W. J.
Wilkins, Jr., J. W. *see* Goldstein, H.
Wilson, T. R. *see* Nyer, W. E.
Wittels, M. C. *see* Crawford, J. H.
Wolfe, G. *see* Bernstien, S.

Yamauchi, H. *see* Feshbach, H.
Young, F. W. *see* Cleland, J. W.

Zinn, W. H. S23, S70
Zinn, W. H. *see* Dietrich, J. R.

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Scientific problems in the development of nuclear power

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The scientific problems in the development of nuclear power divide conveniently into the physical, chemical and metallurgical problems. The physicist is concerned with prediction of the nuclear performance of reactors. He has to be able to predict the critical size and the amounts of fuel and the degree of enrichment required to enable it to operate at the designed heat output. He has further to be able to predict the course of the reactivity—how much heat can be extracted per ton of fuel before the fuel has to be changed. In order to do this he must know a great deal about the interaction of neutrons with the important heavy element isotopes inducing plutonium 240 and 241. He must know how the number of neutrons per fission changes with neutron energy and the relative chances of a neutron being captured with and without producing fission. He must know the chances of neutrons being absorbed by the fission products formed in the fuel elements, and any non-fissile materials used by the metallurgists and engineers. Different data are required for fast and thermal reactors. The physicist must also keep a watchful eye on the safety of reactors—he must be wary lest changes in temperature or operating conditions induce additional reactivity and cause power increases. He must also study the effects of high speed, high intensity neutron bombardment on the properties of important structural materials in the reactor such as graphite and steel.

The chemists and metallurgists have the problem of providing all the nuclear materials—fissile and non-fissile—in a high degree of purity. The chemist must devise processes for separation of fertile materials, fissile materials and fission products. The metallurgist has to understand the effect of radiation on uranium and thorium metal at the operating temperature and has to devise methods of counteracting deleterious effects. To do this he has to study the various possible alloys or cermets bearing in mind the prohibitions of the physicist who dislikes neutron absorbing materials and the chemist who dislikes the job of processing highly alloyed metals.

The chemists and metallurgists have also to study the compatibility of all the materials used with the coolants specified by the engineer. Thus the reaction of graphite with carbon-dioxide gas at 400° C in the presence of reactor radiation must be measured; the compatibility of liquid sodium with zirconium and stainless steel at 550° C must be determined for a reactor of the sodium–graphite type; solutions of uranyl sulphate in water must be compatible with structural materials used in homogeneous reactors. All these problems require prolonged study in test rigs or reactor experiments.

The scientist must also plan ahead, for any new type of reactor is likely to take at least 10 years to come into full scale use from the commencement of work. During this period he has to carry out research and development on any new materials which may be required such as heavy water, thorium, zirconium and beryllium. Work has to be started many years before full scale use can be certain.

The crystal ball is never more required than in the development of atomic energy.

INTRODUCTION

We have now reached a stage in the development of nuclear power where the first large scale prototype stations are being built in at least four countries. In the United Kingdom the first reactor of the Calder Hall nuclear power station reached criticality on May 22nd, 1956. The power station will be formally opened by Her Majesty, the Queen, in October of this year and we expect to be delivering over 65 000 kW of electricity to the national grid early in 1957.

The Atomic Energy Authority are building three further stations of the Calder Hall type. Simultaneously with these, four industrial organizations are designing improved versions of the Calder Hall power station for the Electricity Authorities of the British Isles. Tenders for these stations are due towards the end of 1956 and the Electricity Authorities in England and Scotland will, according to the plan set out in the Government's white paper, place contracts for two stations, as a first instalment of the Government ten-year plan for the development of nuclear power. The output of the first C.E.A. (Central Electricity Authority) stations is expected to be much larger than that of Calder Hall.

Although the design of the first C.E.A. stations will be a substantial advance over Calder Hall there is still a great deal of scope for research and development leading to further improvement of the gas-cooled graphite moderated reactor type of nuclear power station. Later types of nuclear power stations using other kinds of reactors present a still wider range of scientific problems. It is proposed to investigate eight types in this country.

These problems divide conveniently into those of reactor physics, solid state physics, and chemistry, and I will discuss them in that order.

REACTOR PHYSICS PROBLEMS

Core structure. The first problem to be considered by a reactor physicist in designing a new type of reactor is to determine the optimum parameters of the core: lattice pitch, rod diameter and size. He first assembles or measures all the relevant nuclear data. These include the fission, absorption, scattering cross-sections of atomic nuclei used in all the reactor core materials; and the number of neutrons emitted

in fission per neutron absorbed by a fissile nucleus—all as a function of neutron energy.

The second step is to make approximate calculations of core parameters to indicate the range of parameters likely to be of interest. After this exponential experiment, assemblies or zero energy critical assemblies are made to establish the general nuclear characteristics of the reactor system. The data from these experiments enables the methods of calculation to be refined and it is then possible to combine the nuclear calculations of the core together with heat transfer and economic analysis to design the best reactor. After this the design may be finally checked by a further zero energy experiment. In particular the effects of engineering deviations from the ideal nuclear design are determined.

This procedure applied to the gas cooled graphite moderated reactor led to a lattice pitch of about 8 in. and a uranium rod diameter of about 1 in.

An important parameter of the core is the conversion factor of ^{235}U to plutonium since this is one of the most important factors determining long term reactivity changes. A typical neutron balance sheet of a good gas-cooled graphite reactor is shown in the table. You will see from this the neutron losses which are important in determining the conversion factor, which is about 0·85.

Neutron balance for PIPPA type reactor

Yield ^{235}U and ^{238}U	2·10
Absorption:	
^{235}U	1·00
^{238}U	0·85
graphite	0·12
parasitic (i.e. sheaths, xenon and control)	0·08
Leakage	0·05
Absorption and leakage	2·10

Reactivity changes. The fuel costs in power reactors are determined by the cost of uranium, the cost of fabricating fuel elements, the burn-up which can be achieved per cycle and the value of the spent fuel. The achievable burn-up depends on the course of the reactivity changes. The general course of these changes is shown in Fig. 1. There is an early drop of reactivity due mainly to the formation of ^{149}Sm followed by a rise due to the conversion of ^{235}U to ^{239}Pu. The greater the conversion factor the greater is the increase of reactivity.

As the burn-up proceeds past 1000 MWD/t the conversion of plutonium to higher isotopes, ^{240}Pu and ^{241}Pu, becomes important. ^{240}Pu is not fissile by thermal neutrons but is converted to form ^{241}Pu with an absorption cross-section of about 1200 barns. ^{241}Pu has a fission cross-section of about 1000 barns and a cross-section for capture to form ^{242}Pu of roughly 500 barns for thermal neutrons. These cross-sections are not yet known accurately enough; so far they have been determined at Harwell by using plutonium with up to 50% of ^{240}Pu. Fig. 2 shows the isotopic constitution after burn-up of 3000 MWD/t. We have now built an electromagnetic separator for producing separated isotopes of plutonium and expect within the next year to reduce the uncertainties in the values of these constants.

Another important factor in determining long term reactivity changes is the effect of fission products of long half lives. We do not know these cross-sections with sufficient accuracy and are now starting an experimental programme to improve our knowledge. We propose as a first step to extract mixed fission products from a fuel rod of a plutonium alloy which has been heavily irradiated in the Chalk River reactor. We shall then make measurements on bulk fission products. Our chemists will also separate some

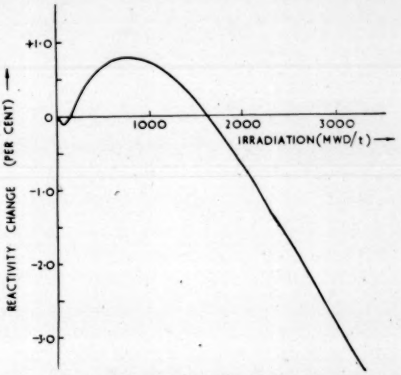

Fig. 1. Isotopic composition of irradiated natural uranium in PIPPA type reactor

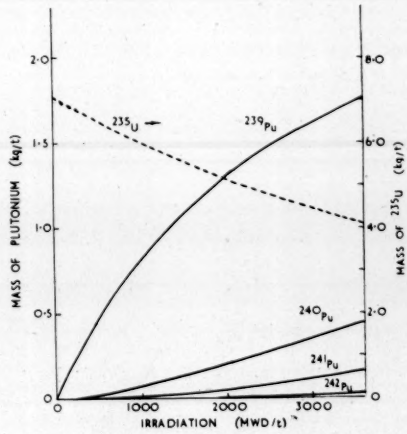

Fig. 2. Reactivity variation with irradiation for a uniformly irradiated PIPPA type reactor

of the more important fission products and cross-section measurements on these will be made. We are hoping to arrange a collaboration programme with the Oakridge National Laboratory on this work.

A programme of this kind requires special facilities and involves a great deal of work. Even to cut up an irradiated

plutonium rod into sections having activities of thousands of curies requires good remote handling devices, whilst the cell for separation of the fission products has to be heavily shielded and to be equipped with remote handling. Facilities of this kind to work with 1000 curies of activity may cost £50 000. The operation of reactors at temperatures adequate to produce power requires a knowledge of nuclear constants at neutron energies higher than thermal. The time of flight methods using fast choppers to interrupt the neutron beams from reactors and the pulsed linear accelerator neutron source are well known, and typical results are illustrated in Fig. 3. We expect to improve the accuracy of our measurements and to extend the range of these by two new developments at Harwell. The first will make use of a neutron beam from the DIDO reactor which is expected to come into use in the autumn of this year. This will give us 40 times higher intensity than we have had from BEPO. A new fast chopper operating at 48 000 r.p.m. is being built for this work and neutron flight path of 100 m will be used. Using a beam of tens of millimetres cross-section and samples of about 100 mg we should be able to get resonance parameters up to about 100 V energy. We are also building a more powerful electron linear accelerator. This will have an electron beam current of 1 A in the pulse and an electron energy of 28 MeV with a pulse width of 1/5 μs and repetition frequency of 400 c/s. The neutron intensity from a heavy target will be about 100 times that available from our present 15 MeV linear accelerator and will be 5×10^{16} n/s in the pulse. We are also planning to use a subcritical assembly of fissile material as a neutron booster to increase the intensity by a further factor of 10.

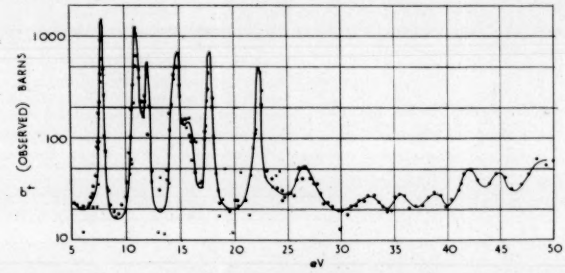

Fig. 3. Cross-sections for ^{239}Pu

■ fast chopper 1·20% ^{240}Pu, 0·08 ^{241}Pu. • linear accelerator ~4% ^{240}Pu, ~0·3% ^{241}Pu.

Another important factor in determining the burn-up in a particular reactor is the cycle of fuel element change which is adopted. The ideal method is to discharge the fuel elements quasi-continuously. This can be achieved if the reactor design provides for charge and discharge to be carried out whilst the reactor is operating.

Since the irradiation of the fuel in the central zones is more intense these are the first to be discharged followed by discharge of zones proceeding outwards. In general, burn-up can proceed until the area below the reactivity curve equals the area above the curve in the early part of the irradiation.

When continuous charge and discharge is not possible operational procedure may lead to discharge by zones, so that the maximum irradiation of individual fuel elements may vary appreciably. So the average burn-up will be less.

With our present inadequate knowledge of nuclear constants estimates of burn-up are uncertain by perhaps 500 MWD/t. The achievable burn-up depends on the neutron economy and particularly on the initial conversion factor. A graphite moderated reactor of the type likely to be used in future power stations is likely to achieve a burn-up of 2500–3000 MWD/t.

An important corollary to this is that slight enrichment of the fuel of graphite moderated reactors is likely to be economical. Comparatively small enrichment to increase the proportion of ^{235}U by 3 or 4% will increase reactivity and allow the burn-up to be increased. The cost of this slight enrichment may be estimated by taking the United States Atomic Energy Commission's price of ^{235}U as a basis. A 3% enrichment requires 215 g of ^{235}U per ton of metal, so that the added cost at $25 000/kg is about £1900 per ton of metal. This would add about 10% to the cost of fuel elements. If the additional burn-up exceeds this increased cost of 10% appreciably, slight enrichment would be advantageous.

Additional burn-up of fissile material in thermal reactors should be achievable by recycling the plutonium together with the greater part of the depleted uranium and feeding into the system a quantity of natural uranium to make up for the ^{235}U burnt.

In the equilibrium condition the proportion of the different fissile isotopes in the reactor stays approximately constant since as much plutonium is produced as is destroyed by fission. The equilibrium state of a recycling reactor operated to recycle fuel in this way has been considered by Fenning and Syrett. Their analysis shows how the overall burn-up achieved depends strongly on the degree of initial enrichment of the fuel, on the conversion factor and on the η value, i.e. the number of neutrons produced per fissile atom destroyed. Since increasing reactor temperature decreases η markedly in the plutonium–uranium cycle this can have an important effect in reducing uranium utilization. Figs. 4 and 5 show a typical curve illustrating the effects of varying η.

We expect that a gas-cooled graphite moderated reactor with a conversion factor of 0·8 and an operating temperature of 400° C could achieve an overall burn-up of the order of 10 000 MWD/t.

The burn-up likely to be achieved in an enriched fuel reactor such as the sodium graphite reactor which we are studying is likely to be markedly less, especially if the reactor is operated at temperatures of about 500° C, for this might

lead to η falling to 1·65–1·70. The value of the conversion factor will, of course, depend greatly on the design and particularly on how much neutron absorbing material such as zirconium is introduced into the core for graphite canning. It will also depend markedly on the reactor size and output. Until design parameters have been specified we cannot therefore predict burn-up accurately. Nevertheless at this stage of investigation it appears that the sodium graphite reactor should be operated on the ^{233}U thorium cycle if good burn-up is to be achieved.

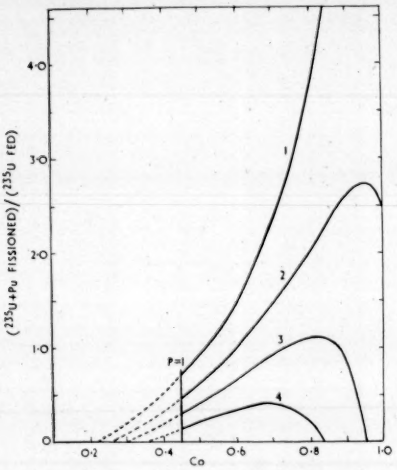

Fig. 4. Variation of uranium utilization with η_{49} (after Fenning and Syrett)

$E_f = 1$, $E_0 = 1{\cdot}25$, $\varepsilon/(1 + K^2L_2^2) = 1$.

Curve	η_{49}
1	2·00
2	1·85 (~ 400° K)
3	1·75 (~ 600° K)
4	1·65 (~ 900° K)

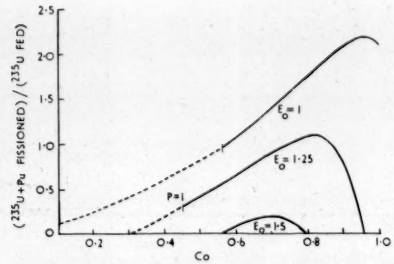

Fig. 5. Variation of uranium utilization with initial enrichment and conversion factor (after Fenning and Syrett)

$E_f = 1$, $\eta_{49} = 1{\cdot}75$, $\varepsilon/(1 + K^2L_S^2) = 1$.

Problems of fast reactors. The problems of reactors using the upper end of the fission neutron spectron are completely different from those of thermal reactors.

The computation of critical core sizes by purely mathematical methods at first led in the U.K. to results which were uncertain to $\pm 25\%$. Since that time we have constructed two zero energy reactors ZEPHYR and ZEUS fuelled by plutonium and ^{235}U respectively and our knowledge of the behaviour of fast reactors has greatly increased.

One of the big uncertainties was the way in which the fission neutron spectron is degraded by inelastic scattering as we go outwards in radius through the core and blanket. A typical example of the measured results is shown in Fig. 6.

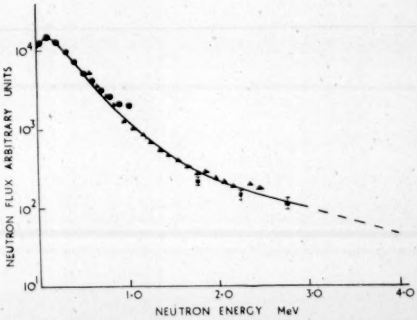

Fig. 6. Neutron spectrum in envelope of ZEPHYR: radius of 28 cm from centre of core (after Holmes and others)

- ● ^3He (n.p.) proportional counter
- ▲ hydrogen recoil, proportional counter
- ■ hydrogen recoil, photographic plates

Important measurements have also been made on the variation of the fission rates throughout the core and blanket, in plutonium, ^{235}U and ^{238}U.* It was found that the fission of ^{238}U contributes in an important way to the neutron economy of the reactor. The overall result was that in the uncooled, under-moderated ZEPHYR, breeding gains of over two were obtained. In a future high-powered plutonium fast reactor, the achieved breeding gains are likely to be between 1·6 and 1·7, which is very handsome. ZEPHYR work has recently shown that with a plutonium core and thorium blanket, 1·6 atoms of ^{233}U are formed for each plutonium atom destroyed.

* See p. S 32 of this Supplement.

PROBLEMS OF SOLID STATE PHYSICS

Problems of solid state physics or metallurgy in nuclear reactors are of even more importance than those of nuclear physics, for the most difficult problem of all is the development of a reliable fuel element.*

The first group of problems is concerned with the radiation stability of fuel elements. The problems vary depending on whether we use metal or oxide for the fuel elements and on the operating temperature.

The first commercial graphite moderated gas-cooled reactors will operate with fuel element surface temperatures somewhat over 400° C and central metal temperatures below the U $\alpha - \beta$ change point of 665° C. Sodium graphite reactors and fast reactors will probably have metal surface temperatures of 500° C, whilst future reactors depending on cermet or oxide fuel elements may operate at much higher temperatures.

The study of radiation damage in any new set of circumstances may require a total effort of 200 man years. A good deal of this effort goes into the preparation of specimens having different alloy constitutions and different heat treatments. Loops have to be prepared for containing and cooling the specimens in a testing reactor. These loops have to be provided with thermostatically controlled temperature conditions.

After the irradiation the specimens are highly active and must be examined by remote viewing behind thick concrete walls and handled by remote control. For this purpose television has been used successfully at Harwell. Fig. 7 shows

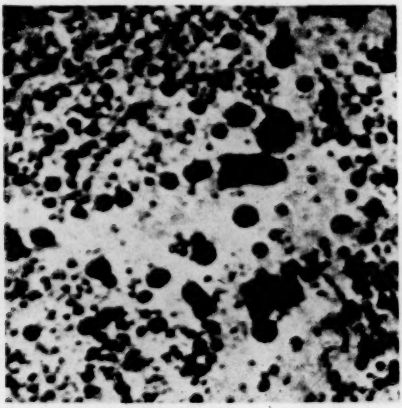

Fig. 7. Specimen having 13% decrease in density on irradiation

examples of photographs taken by a television tube showing irradiated specimens.

The low temperature radiation damage can lead to severe distortion of uranium.† We are still very far from understanding this phenomenon though it undoubtedly results from the complicated orthorhombic crystal structure of uranium. The large-scale effects can cause a uranium rod to stretch in length or to shrink several inches depending on crystal orientation, as illustrated in Fig. 8. A single crystal of uranium changes its shape markedly under radiation, just

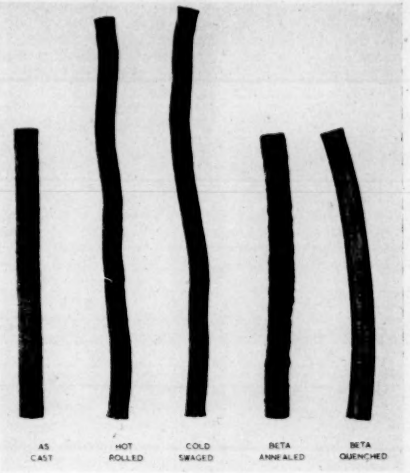

Fig. 8. Uranium bars, initially of equal length, after 850 cycles between 600 and 50° C

as though the uranium atoms flowed through the crystal stimulated by the local heating due to the high-speed fission fragments. The immediate solution for uranium fuel elements to be operated at temperatures of the order of 400° C is to heat treat the metal so as to reduce the grain size and to obtain random orientation of the crystals in wrought bars. This very greatly reduces the distortion of the uranium rods and it is now felt that this distortion will not be the limiting factor in the burn-up achieved in graphite reactors of the Calder or C.E.A. type.

At the higher temperatures which are likely to be characteristic of the fast reactor or sodium graphite reactor the uranium is more plastic and we now encounter a different problem due to the pressure exerted by the fission gases. They appear to migrate to particular localities and to form high pressure gas pockets which cause bulk swelling of the metal. We are far from understanding what decides the number and distribution of these pockets. If the metal is unrestrained the increase of volume may be as much as 40% at 800° C. An immediate experimental answer to this form of distortion may be to provide a strong sheath. In this way the growth may perhaps be kept down to about 5%. The full scale fast reactor may well have to use a liquid fuel.

Another important problem is the effect of irradiation on mechanical strength and creep. At the Geneva Conference one of the Russian papers pointed out that under irradiation creep increased in some circumstances by a factor of the order of 1·5 to 2. Dr. Cottrell has investigated this effect

* See p. S 91 of this Supplement.
† See p. S 43 of this Supplement.

by irradiating loaded uranium springs in the Harwell BEPO. He finds that during irradiation at 100° C the metal can creep slowly (0·1% in a year) under stresses that are only about one-hundredth of the normal creep stress of the metal at that temperature. This could be important in particular reactor designs where the uranium metal was heavily stressed.

Similar problems arise, though not in such an acute form in the case of most metals used under strain in reactors. An obvious example is the possible effect of radiation on the breaking strength of the pressure drum containing the reactors.

The effect of radiation on solid moderators is another important problem. In addition the most striking effect has been found in the irradiation of graphite. Graphite crystals grow in one direction and contract in another owing to the formation of interstitial atoms by neutron impact. The result is that bulk graphite grows by 0·05% during a year in a flux of 10^{13} n cm^{-2} s^{-1} at a temperature of 150° C. Some

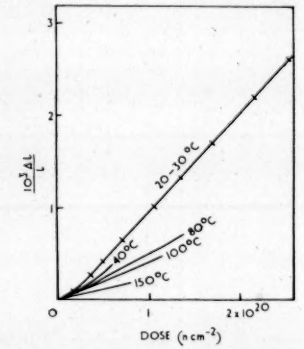

Fig. 9. Growth perpendicular to extrusion direction at different irradiation temperatures (after Kinchin)

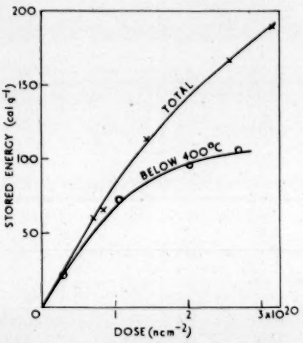

Fig. 10. Total stored energy and energy released below about 400° C, irradiation temperature 30° C (after Kinchin)

typical effects are shown in Fig. 9. At the same time there are striking changes in the physical properties such as thermal conductivity, which is reduced by a factor of 7 under such conditions. The effects depend strongly on temperature since at high temperatures the damage anneals out as it is produced.

Along with this so-called "Wigner growth" goes a storage of energy due to interstitial atoms as illustrated in Fig. 10. At a temperature of 100° C and an irradiation of 1 year at 10^{13} n cm^{-2} s^{-1} the stored energy is 110 cal/g; at 30° C it is 190 cal/g. A large part of this energy can be released by heating the graphite above the irradiation temperature (a typical example is shown in Fig. 11). The graphite specimen will then show a quite spectacular spontaneous heating.

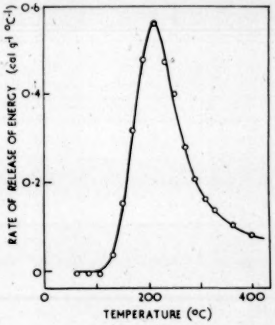

Fig. 11. Rate of release of energy. Linear rise of temperature 20° C/min. Irradiation $\sim 2 \times 10^{20}$ n cm^{-2} at 50° C

We still have a great deal to learn about the storage mechanism and behaviour at high temperature—particularly about the reason why part of the stored energy does *not* cancel out.

Different problems may arise if and when we use beryllium or beryllia moderators at high temperatures. We may be concerned here with effects arising from the formation of helium by (η, α) reactions. This may build up gas pressure and lead to swelling and distortion.

CHEMICAL PROBLEMS

As a physicist I speak with great diffidence about the chemical problems of nuclear power development; Dr. Spence will speak on this subject during the Conference.* I can emphasize their importance by telling you that our chemists and chemical engineers and their problems are as numerous as our physicists and their problems.

The role of the chemist in the development of nuclear power is illustrated in Fig. 12, which shows the flow of uranium and fissile material in a nuclear power system. In the initial stages uranium has to be separated from its ores and concentrated into very pure uranium oxide or uranium fluoride. Chemical research has shown that after reacting the ores with common reagents, ion exchange resins can be used to separate the uranium from the weak solutions in a

* See page S 88 of this Supplement.

very simple and elegant way. This now forms an important part of the extraction process of some plants. In other plants solvent extraction processes have been used.

The next stage of the flow sheet is the reduction of uranium metal. The classical methods have reduced uranium fluoride by calcium or magnesium and these processes have now reached a high efficiency.

The next group of problems arise in the reactor itself.

One important group of problems is concerned with the compatibility of the materials used in reactors, particularly the compatibility of coolants with fuels and moderators and canning materials.

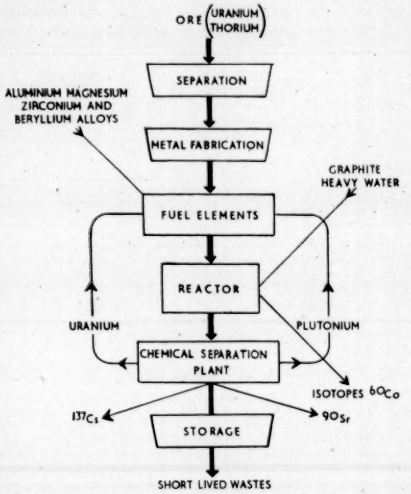

Fig. 12. Chart of flow of materials

In gas-cooled graphite moderated reactors the principal compatibility problem is that of the carbon dioxide coolant with the graphite moderator. The reaction leads to the formation of carbon monoxide and an equilibrium concentration is established. The reaction could lead to some mass transfer of carbon from one part of the reactor and its cooling circuit to another. The reaction rate increases a thousandfold with radiation at temperatures of 300–400° C and a loop has been operating for some time in BEPO to study the magnitude of the radiation effect. The results have shown that at the graphite temperatures we may expect at Calder Hall, and in the future C.E.A. reactors, the effects are not likely to be significant but they might prove to be a limiting factor in raising the temperature of the reactor core well beyond 400° C. In order to make higher temperatures possible with carbon dioxide as a coolant it would probably be necessary to find means of reducing the porosity of the graphite.

In water-moderated reactors the principal problem is the compatibility of the hot water with the uranium fuel, for if a fuel element sheath develops a leak it must not be possible for a rapid attack of the fuel to take place. Experiment has shown that unalloyed uranium is attacked far too rapidly to be tolerated and if metal fuel elements are used it is necessary to resort to alloying to reduce the corrosion rate. This increases the neutron absorption markedly and a preferable alternative for commercial reactors is to use sintered uranium oxide fuel elements, since they are very resistant to attacks by hot water.

The hot water must also be compatible with the fuel element sheath, and to achieve this Zircalloy—an alloy of zirconium—has been developed in America. Research is proceeding at Harwell on a range of aluminium alloys which might withstand the conditions in water reactors. If successful, these alloys would lead to a reduction in fuel element costs.

Compatibility must also be achieved with the pressure vessel and external circuit material. An obvious but expensive method is to use stainless steel throughout or at least stainless steel linings. However, stainless steel is difficult to decontaminate from fission products and is liable to stress corrosion and it may well be possible as a result of loop tests now proceeding and adequate control of water conditions to use mild steel.

In sodium graphite reactors the hot sodium is compatible with uranium metal fuel, so one compatibility problem is removed. On the other hand the sodium must not be allowed to penetrate the pores of the graphite or too much reactivity would be lost. So the sodium must be kept away from it. One solution which has been proposed is to can the graphite in hafnium free zirconium sheet.

We now strike the unpleasant fact that zirconium is a very good "getter" for oxygen and so will take up any oxygen leaking into the system. Oxide leads to embrittlement and so the onus is thrown back on the designer to devise methods of keeping the oxygen content of the sodium below 10 parts per million. This difficulty may well force the designer to use a calandria design.

Compatibility problems are still more acute in the aqueous homogeneous reactor for it has been found that the corrosion rate of the zirconium vessel holding the core fluid—uranyl sulphate—increases strongly with radiation intensity. A search has therefore to be made for corrosion inhibitors or other materials have to be sought which are more corrosion resistant in the presence of radiation.

In solid fuel reactors, another group of chemical problems arises from the need to recycle fuel if overall burn-ups of the order of 10000–20000 MWD/t are to be achieved in future nuclear power systems. The well-proven aqueous solvent extraction processes are likely to be used and in large scale plants they are likely to achieve low separation costs—of the order of £1000 per ton of uranium metal processed. This has to be compared with an initial fuel element cost of £15000–£20000 per ton. The separated uranium and plutonium will now have to be refabricated under conditions under which the plutonium is contained in closed boxes, because of its toxicity. Here is a good opening for automatic remote controlled fabrication. We do not yet know the costs of refabrication under these conditions, but development work is proceeding to determine this.

Quite different chemical processing methods will be required for the breeder reactors. The fast breeder will possibly require a large number of recycles in the life of its expensive fuel owing to the severe radiation damage. Aqueous processing methods require the fuel to cool for 100 days or so to allow the radiation level to decrease. These two factors together will probably lead to unacceptably high fuel investment and processing costs so that chemists are

now working on pyro-metallurgical processes—which essentially consist of melting the fuel elements, skimming off the waste products and re-forming. This is likely to be a lengthy development job.

In thermal breeders such as the aqueous homogeneous reactor and the liquid metal fuelled reactor, continuous processing of the core is possible in principle and work has been proceeding on this for some time. The fission gases, xenon and krypton, can be extracted and their removal

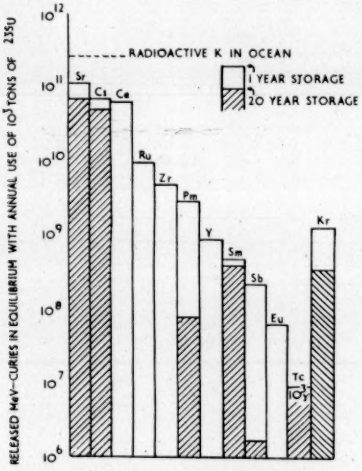

Fig. 13. Decay of activity of isotopes after removal of ^{137}Cs and ^{90}Sr

substantially improves the neutron economy of the system. Other fission products forming solid particles can be removed by a cyclone.

The last section of the flow diagram showed the treatment and disposal of the radioactive waste products which are separated in the chemical plant.

When the U.K. nuclear power stations reach an output of 20000 megawatts they will be producing about 6 tons a year of radioactive fission products. This is not a large volume—certainly it is small compared with the volume of ashes of present U.K. power stations. We can therefore deal with the problem by evaporation of the liquids and storage in large stainless steel tanks of the type we now use at Windscale. Papers at the Geneva Conference, particularly from Hanford, showed that storage would contribute only one or two per cent to the cost of nuclear power. I wish to emphasize that we do *not* put concentrated waste products in the sea, but only plant washings of weak activity—100 c.p.d., which are diluted by dispersion to a volume of 10^{13} cm^3 in about a day.

The waste products do, however, contain radioactive isotopes, which are potentially valuable. ^{137}Cs which emits 600 kV gamma rays and has a half-life of 27 years is an important example.

Fig. 13, taken from Dr. Glueckauf's Geneva paper shows how the activity of the different isotopes decay after ^{137}Cs and ^{90}Sr are separated and treated separately.

The Windscale research laboratories are now separating ^{137}Cs on a pilot plant scale and the product is soon going to be used to provide kilocurie sources for radiotherapy in hospitals. Caesium might well in time replace 200 kV X-rays for therapeutic treatment.

Other potential applications are to industrial processes and here sources of the order of a million curies might be available by the 1960's.

The development of these applications requires a good deal of research work by chemists and biologists. We are now building up as a close satellite of Harwell, the Wantage Radiation Laboratory which will be equipped with powerful radiation sources, and where such research will be carried out. We are already collaborating closely with industry in the application of our radiation sources to their problems. When a specific application is shown to be worth while industry is already equipping itself with suitable radiation sources.

CONCLUSION

You will see that the development of nuclear power requires an intensive programme of research and development continuing over at least the next two decades.

At the present time the Atomic Energy Authority has about 1000 qualified scientists and technologists working on this development—which is a little over 2% of the total research and development staff of the country, and is about one-tenth of the effort in the United States.

In view of the magnitude of the nuclear power programme and the fact that it is basic to our whole economy this does not seem to be an undue proportion of our resources.

The effort is now being supplemented by industry in the design of nuclear power stations and as the programme builds up, their share will increase. Given this additional effort there is little doubt that nuclear power will make a substantial contribution to our energy resources by 1965 and that its contribution will grow rapidly in the second decade.

SESSION 2. LECTURE

Chairman: SIR JOHN COCKCROFT, K.C.B., C.B.E., D.Sc., F.Inst.P., F.R.S. (*President*)

The physics of advanced reactors

By J. R. DIETRICH, Ph.D., Argonne National Laboratory, Lemont, Illinois, U.S.A.

This lecture is an attempt to characterize the various promising types of solid fuel power reactors in terms of the neutron-physics characteristics of their moderators, coolants, and fuel cycles.

The neutron-physics design objectives which have the greatest effect on power cost are:

(i) use of a fertile isotope (uranium 238 or thorium 232) as a major source of energy; (ii) internal conversion ratio high enough to allow long fuel element life; (iii) safety characteristics favourable enough to avoid remote location and high containment cost.

These objectives are not restrictive enough to define the "best" reactor type, but they are the major factors in determining the composition and geometric arrangement of specific reactor designs once the type has been chosen.

The thermal reactors which are at present being designed or constructed utilize uranium 235 as the initial fissile isotope, and there is a strong incentive to utilize ratios of fissile to fertile material which depart as little as possible from the "natural" value. With this additional design restriction, the lattice characteristics appropriate to the various moderator-coolant combinations are still more sharply defined. Examples are drawn from the current group of reactor designs to illustrate these characteristics. The possible consequences of a greater latitude in the choice of fissile-fertile ratio are discussed, and the relative characteristics of the possible fuel cycles are considered.

The characteristics of fast reactors fit into a quite different pattern. The possible conversion ratio is high on either fuel cycle, and the reactivity changes associated with fuel burn-up are relatively low simply because of the very large fissile-material content of the reactor. The requirement that the average neutron energy be kept high is a rather restrictive one. It appears now that the range of variation in fast reactor design will be less than that in the thermal reactor field. The characteristics of typical fast reactors are presented, their special problems are discussed, and some recent experimental results are quoted.

INTRODUCTION

The considerations in reactor performance which strongly influence the neutron physics of the reactor design may be stated as follows:

(*a*) A large fraction of the reactor energy output should be derived from one of the fertile materials ^{238}U or ^{232}Th.

(*b*) The economic advantages are great for reactors which have internal conversion ratios sufficiently high that the life of fuel elements is determined by fission damage rather than by reactivity loss.

(*c*) A real economic advantage results from safety in the reactor design.

In addition to these objectives which apply generally, it is usually found that in any given type of reactor the economic performance is improved by an increase in power density.

The above considerations are by no means adequate criteria for judging the relative merit of different reactor types. They do represent the strongest considerations affecting the design of a reactor of any given type, and it is possible to characterize roughly the various reactor types in terms of the way they meet these objectives.

So far as reactor physics is concerned, items (*a*) and (*b*) above specify that the reactor should have a high conversion ratio. At the moment it is not possible to assign necessary values for the conversion ratio or to state quantitatively the worth of a given conversion ratio. This difficulty arises because a number of possible fuel cycles exist; relative costs of fuel, reprocessing, and fabrication are not available; and the degree to which fission damage limits fuel element life is variable. The following fuel cycles are among those possible:

(*a*) *Once-through use of natural uranium without plutonium recycle.* If any appreciable consumption of ^{238}U is to be achieved on this basis, the atom burn-up per fuel element must be quite high. Fig. 1 shows the relationship of conversion ratio to atom per cent burnup in uranium fuel elements for some postulated modes of operation.[1]

(*b*) *Natural uranium feed with plutonium recycle.* Papers by Lewis[2] and by Dunworth[3] have pointed out that significant fractions of ^{238}U can be consumed if the conversion ratio is in the neighbourhood of 0·8.

(*c*) *Recycle of plutonium in a fast reactor-thermal reactor system.*

(*d*) *Thorium–^{233}U cycle with ^{233}U recycle.*

In any of the above cycles long fuel element life is advantageous from the standpoint of reducing reprocessing and fabrication costs. As rough figures of merit it might be said that in a uranium–plutonium system, the minimum conversion ratio of interest is about 0·7, that a ratio of 0·8 makes possible the consumption of a few per cent of the ^{238}U, and that any improvement on 0·8 will have a large advantage both economically and in the conservation of the uranium resource.

Many current plans for power reactors envisage the construction of a pilot plant of lower power output than that expected ultimately to be economic. These pilot plants are generally designed to prove the uncertain design features of the reactor type, and seldom have neutron physics characteristics typical of the full scale reactor. For a general understanding of the characteristics and limitations of the various reactor types it is therefore more profitable to consider the basic physical principles rather than the detailed neutron physics of proposed pilot plants. If the discussion is limited to solid-fuel reactors such a consideration can be centred around the type of moderator used, the fast reactor being considered as the limiting case of no moderator. It is useful to take as the point of departure the natural uranium

reactor, to examine the limitations placed on it by the use of natural uranium, and to observe the way in which enrichment of the fuel makes possible improvements in performance as well as making possible the use of entirely different reactor types. In addition to these general considerations some of the specific characteristics of reactor types under development at Argonne National Laboratory will be discussed briefly.

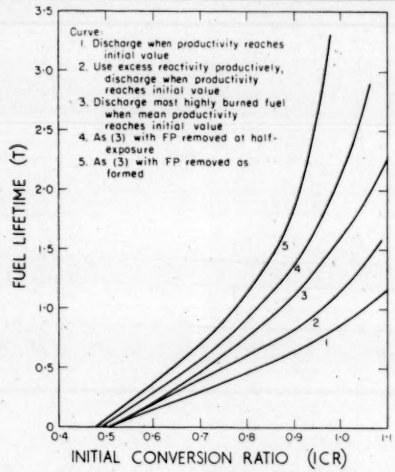

Fig. 1. Fuel lifetime as limited by reactivity loss under various operating methods, plotted as fissions/^{235}U atom in feed (T) *v.* initial conversion ratio (ICR), for ^{235}U–^{238}U systems. The productivity is defined as the difference in rates of neutron production and loss per unit of flux time

CRITICALITY OF NATURAL-URANIUM REACTORS

A list of symbols and definitions will be found in the appendix

In designing the natural uranium reactor the first concern is usually to provide that the reactor be critical under all desired operating conditions. The requirements for criticality can be expressed approximately by the elementary criticality equation

$$\frac{k}{1 + M^2 B_g^2} = 1 \qquad (1)$$

Here

k = number of second generation fission neutrons produced/number of first generation neutrons absorbed in the reactor;

$\dfrac{1}{1 + M^2 B_g^2}$ = fraction of neutrons born which are absorbed in the reactor (i.e. which do not leak out of the reactor);

k, then, characterizes the process of neutron reproduction within the reactor, while the term $M^2 B^2$ characterizes the loss of neutrons from the reactor by leakage. M^2, the migration area, is a property of the reactor composition, and is proportional to the mean square distance that would be travelled by fission neutrons between birth by fission and death by absorption if the reactor were infinite in extent. The geometrical buckling B_g^2 is the solution of the partial differential equation for the neutron flux ϕ in the reactor, with proper boundary conditions:

$$\nabla^2 \phi + B_g^2 \phi = 0 \qquad (2)$$

Thus B_g^2 is a property of the reactor geometry alone.

It is customary to define a material buckling B_m^2 for the reactor material alone, defined by the equation:

$$B_m^2 = (k - 1)/M^2 \qquad (3)$$

Note that while equations (1) and (3) are formally identical, they express two quite different relationships. Equation (1) is a criticality condition; equation (3) defines a property of the reactor composition, regardless of criticality. The criticality condition in terms of B_m^2 is:

$$B_m^2 = B_g^2 \qquad (3a)$$

B_m^2 expresses, in a single number, the potentiality of a given reactor composition for criticality. If $B_m^2 > 0$ a critical reactor can be had by making the reactor dimensions large enough to satisfy equation (3*a*). In general B_g^2 is a sum of terms involving the reciprocals of the squares of the dimensions of the reactor [e.g. $B^2 = (\pi/H)^2 + (2{\cdot}405/R)^2$ for a cylinder of height H and radius R].

The moderators whose nuclear properties make possible $B_m^2 > 0$ with natural uranium are D_2O, graphite, beryllium, and BeO. Of these, only D_2O and graphite have been seriously considered for power reactor use. The important neutron properties of these two materials are tabulated in Table 1.

The quantities Σ_a, Σ_s, and $\sqrt{(\xi \Sigma_s / \Sigma_a)}$ are related to the ability of the moderator to sustain a high multiplication factor k in the reactor. Their significance can be appreciated by considering the usual four-factor formula for k, as it would apply to a reactor composed entirely of the moderator and natural uranium.

$$k = \eta \epsilon p f \qquad (4)$$

where η is the regeneration factor, ϵ the fast-fission reactor, p the resonance escape probability, and f the thermal utilization. For purposes of approximation, η and ϵ can be considered properties of the fuel alone. To simplify matters further, disadvantage factors will be ignored, and the resonance integral of ^{238}U in the fuel lump will be considered a constant,

Table 1. *Comparison of properties of moderators*

Moderator	Σ_a cm^{-1}	$\xi\Sigma_s$ cm^{-1}	λ_{tr}(thermal) cm	τ cm^2	$\sqrt{(\xi\Sigma_s)/\Sigma_a}$
Graphite ($\rho = 1{\cdot}60$)	$3{\cdot}53 \times 10^{-4}$	0·0611	3·14	364	13
D_2O (room temperature)	$1{\cdot}05 \times 10^{-4}$	0·175	2·65	125	41
H_2O (room temperature)	0·0221	1·31	0·48	33	8

independent of lump geometry. Equation (4) can then be expanded:

$$k = \eta\epsilon \left\{ \exp\left[-\frac{V_u}{V_m} \frac{\int_{res} \Sigma_{28}(eff)dE/E}{\xi\Sigma_s} \right] \right\} \left(\frac{V_u\Sigma_{au}}{V_u\Sigma_{au} + V_m\Sigma_{am}} \right) \quad (5)$$

where V_u and V_m are the volume fractions of uranium and moderator, respectively; Σ_{au} and Σ_{am} are the thermal absorption cross-sections for uranium and moderator, $\xi\Sigma_s$ applies to the moderator, and $\int_{res} \Sigma_{28}(eff)dE/E$ is the effective resonance absorption integral for the ^{238}U as it exists in the reactor. All cross-sections are macroscopic (cm^2 per cm^3 of material).

Giving k the maximum value with respect to V_m/V_u yields the result that k_{max} is attained when p is approximately equal to f (i.e. when the thermal absorption of neutrons by the moderator is about equal to the resonance absorption by ^{238}U). A further result is that the quantity $\sqrt{(\xi\Sigma_s)/\Sigma_a}$ for a moderator is a figure of merit which indicates its efficacy in promoting a high value of k for a given fuel enrichment. The quantity $(\xi\Sigma_s\Sigma_a)^{-\frac{1}{2}}$ is a rough index of the volume ratio of moderator to fuel at which maximum k will be obtained if no absorbers other than fuel and moderator are present. Maximum buckling will usually occur at a lower value of V_m/V_u than that for maximum k because lower values of V_m/V_u result in lower leakage of thermal neutrons.

The remaining items of Table 1 (λ_{tr}, τ) characterize the neutron leakage properties of the moderator. λ_{tr}, along with the average absorption cross-section in the *reactor* (Σ_a) determines the diffusion area (L^2):

$$L^2 = \lambda_{tr}/3\Sigma_a \text{ for dilute systems}$$

The migration area M^2 is the sum of L^2 and the Fermi age (or slowing down area) τ.

Examining Table 1, it is evident that D_2O will promote a considerably higher value of k than will graphite, that lower moderator/uranium ratios will be required for a given resonance escape (higher $\xi\Sigma_s$), and that fast neutron leakage will be considerably smaller in D_2O for a given reactor geometry (smaller τ). Thus it is to be expected that lower moderator/uranium ratios will be used with D_2O than with graphite, and that the attainable values of buckling will be quite considerably higher.

Fig. 2 shows experimental values[4] of B_m^2 for D_2O and graphite-moderated lattices.[5] The moderator/uranium ratio for maximum buckling ranges from 17 to 28 for D_2O, depending on rod size, and would perhaps range from 40 to 60 for graphite. The maximum buckling attained in D_2O is about seven times that in graphite.

The details of the performances of the two moderators are summarized in Table 2 for comparable rod sizes. The factor of about 7 between B_m^2 for D_2O and for graphite arises from a factor of 3·2 attributable to $(k - 1)$ and a factor of 2·3 attributable to M^2. Much of the latter effect may be lost if the reactor is used at high temperature because of the very high volume coefficient of expansion of water relative to that of graphite. If, for example, the D_2O reactor is to be used at 287° C (550° F), the D_2O density will be only 75% of the room-temperature value. The value of k can be maintained at this temperature if the atomic ratio of moderator/uranium

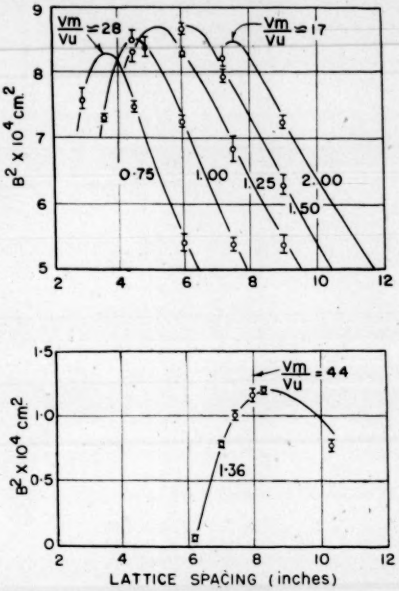

Fig. 2. Bucklings for various lattices: in D_2O (*above*) and in graphite (*below*). The rod diameters in inches are shown on the curves. The D_2O experiments were made with the fuel in aluminium tubes of 0·1 cm wall thickness. The graphite assemblies included a volume of aluminium equal to 4·25 cm^3 per cm length of fuel rod. The holes in the graphite for the fuel slugs were 2·215 cm in diameter. D_2O data from reference 4 (Cohen); Graphite data from reference 5 (Gast)

Table 2. *Comparison of multiplying lattices in three moderators*

Source of data	*Moderator*	*Rod diam. (in.)*	$\frac{V_m}{V_u}$	*Lattice space (in.)*	τ_t	ε	p		k	M^2 cm^2	B^2 cm^{-2}	2·945/B cm
Reference 5	Graphite	1·36	44	8	1·32†	1·03*	0·87*	0·904	1·064	552	$1·2 \times 10^{-4}$	267
Reference 4	D_2O (R.T.)	1·50	19·4	6	1·32†	1·03*	0·93*	0·95	1·207*	240*	$8·65 \times 10^{-4}$	100
Reference 8	H_2O(R.T.) (1·143% U^{235})	0·60	2	1·03	1·53	1·06	0·80	0·89	1·114	32	$48·2 \times 10^{-4}$	42·4

* These values are estimated, since they were not quoted specifically in the original source of data.

† For consistency in comparisons a value of 1·32 is used for η in all natural-U lattices rather than the slightly different values quoted for the original experimental data.

is maintained at the room-temperature value. Such an adjustment, however, will increase M^2 in inverse proportion to the square of the D_2O density—i.e. to 427 cm^2. At the same time, if graphite of higher density than that used in Table 2 is available, the value of M^2 for graphite can be correspondingly decreased. Conversely, the incorporation of large void fraction for gas coolant channels will increase M^2.

The last column of Table 2 is the radius of the minimum-volume critical cylindrical reactor (core plus reflector saving) corresponding to the value of the buckling. These values would be appropriate for low-power research reactors operating at room temperature. Power reactors require additional reactivity to compensate for the fission products ^{135}Xe and ^{149}Sm. In Table 3 the graphite and D_2O reactors of Table 2 are examined in terms of high-power, high-temperature operation. An effective lowering of k by 3·5% has been allowed for xenon and samarium poisoning, and for operational flexibility, and a further loss of 1·5% has been attributed to the effect of high-temperature operation. In addition the moderator/uranium ratio of the D_2O reactor has been raised to 26, and M^2 has been raised to 427 cm^2 to correspond to a reactor designed for, and operated at, 287° C (550° F). The critical sizes have now become much larger.

Table 3. *Graphite and D_2O reactors with reactivity losses due to ^{135}Xe, ^{149}Sm, and temperature*

Moderator	*Rod diam. (in.)*	$\frac{V_m}{V_u}$	k	M^2 *cm²*	B^2 *cm⁻²*	B *cm⁻¹*	$R_{min} = 2{\cdot}945/B$ *cm*
Graphite	1·36	44	1·011	552	$0{\cdot}199 \times 10^{-4}$	$4{\cdot}47 \times 10^{-3}$	659
D_2O 287° C (550° F)	1·50	26	1·147	427	$3{\cdot}45 \times 10^{-4}$	$1{\cdot}86 \times 10^{-2}$	158

Quite obviously an optimum reactor design will achieve better performance than can be demonstrated by the simple approach used here. Nevertheless it is fairly evident that the graphite-moderated power reactor has little reactivity to spare when operating on natural uranium. The D_2O-moderated reactor has some reactivity to spare, particularly if its size is allowed to become large. Difficulties of fabrication and transportation of large pressure vessels have been cited as limitations on the size of water-moderated reactors. If it is assumed that 15 ft is the maximum practical vessel diameter the corresponding buckling is $1{\cdot}71 \times 10^{-4}$. With the properties of Table 3 this reactor would have an excess buckling of $1{\cdot}83 \times 10^{-4}$ cm, or an excess k of 6·5%. It is instructive to consider the possible uses for this excess reactivity, for similar and more extensive benefits can be obtained in any reactor type by the use of fuel enrichment.

Three methods exist for using excess reactivity to increase specific power. They may be used separately or in combinations:

(*a*) *Flux flattening.* k is made lower near the centre of the reactor to give a lower ratio of maximum/average power density, and thus permit higher average power. The decrease in k may be achieved by increasing the proportion of ^{238}U absorption, and thus improving conversion ratio at the same time.

(*b*) *Decrease of moderator/fuel ratio.* This increases heat-transfer surface per unit volume of reactor.

(*c*) *Subdivision of fuel elements.* Large fuel rods are replaced by clusters of small rods or plates. This increases heat-transfer area. The resonance absorption is increased in the D_2O-cooled reactor because the self-shielding of resonances is decreased.

Alternatively, excess reactivity can be used up in useful but absorbing materials. Such materials include alloying materials for the fuel, cladding materials, coolants, and flow tubes. In using up the 6·5% excess reactivity mentioned above, one could allow a 6·5% decrease in f, or an additional absorption equal to 7·2% of the uranium absorption. The average macroscopic thermal absorption cross-section of natural uranium is 0·32 cm^{-1}. The permissible volume fractions of several materials are listed in Table 4. The quantities in column 3 of the Table must be divided by a disadvantage factor having a value probably in the range between 1 and 2, depending on the location of the material with respect to the fuel. It is evident that substantial quantities of materials in the aluminium–zirconium cross-section range can be tolerated, and, indeed, some will be needed, for the quantity of aluminium present during the measurements of Table 2 is insufficient to cover the canning and structural needs of a practical reactor.* The tolerable quantity of zirconium is sufficiently high to suggest the feasibility, from the neutron-physics standpoint, of a pressurized-tube reactor. Such a reactor would not be subject to the pressure vessel limitations previously assumed.

Table 4. *Allowable volumes of parasitic absorbers*

Material	Σ_a	$\frac{cm^3 \text{ of material}}{cm^3 \text{ of } U}$
Al	0·0123	1·9
Zr	0·0068	3·4
Na	0·0114	2·0
H_2O*	0·0244	0·94
Fe	0·19	0·12

* Effect on moderation neglected.

CONVERSION RATIO OF NATURAL-URANIUM REACTORS

One expression for the conversion ratio in a thermal reactor is:

$$R_c = \frac{\Sigma_{28}}{\Sigma_{25}} + \eta_{25}^{\varepsilon} \frac{(1-p)}{1+\tau B^2} \tag{6}$$

$$= \frac{0{\cdot}57}{E} + \eta_{25}^{\varepsilon} \frac{(1-p)}{1+\tau B^2} \tag{7}$$

where η_{25}^{ε} is the regeneration factor for pure ^{235}U and E is the enrichment = (fraction of ^{235}U in fuel)/(fraction of ^{235}U in natural uranium).

Thus the conversion ratio for a natural-uranium reactor can never go below 0·57, and, in fact, because of criticality requirements, will always be substantially above this figure. Equation (7) gives, for the maximum-buckling graphite reactor of Table 2, $R_c = 0{\cdot}82$. For the D_2O reactor in the same table, $R_c = 0{\cdot}69$. This relatively low value for the D_2O case results from the fact that the maximum-buckling

* In this connexion, note that the graphite reactor of Table 3 had an Al/U volume ratio of about 0·5, whereas the D_2O reactor in the table had a ratio of about 0·1 (see Fig. 2).

arrangement results in low resonance absorption by ^{238}U; it has been assumed that the excess neutrons made available by the high value of k are allowed to leak from the reactor. If k is reduced to 1·064 by decreasing the D_2O/uranium ratio the allowable leakage is correspondingly decreased, and the conversion ratio rises to about 0·93 or perhaps a little higher. Since the term $(1 - p)$ is the important variable term of equation (10) it is primarily the moderator/uranium ratio which determines the conversion ratio of a natural-uranium reactor: it is this ratio which determines how many neutrons are absorbed in ^{238}U resonances; for those absorbed thermally in the fuel the ratio of ^{238}U to ^{235}U absorptions is fixed; whether those which escape absorption in the fuel altogether are allowed to leak or are absorbed parasitically has a relatively minor effect on conversion ratio.

LIMITATIONS ON ENRICHMENT

For a given conversion ratio, enrichment does not have a large effect on the reactivity change associated with a given (large) fuel irradiation (Fig. 3). However, the percentage

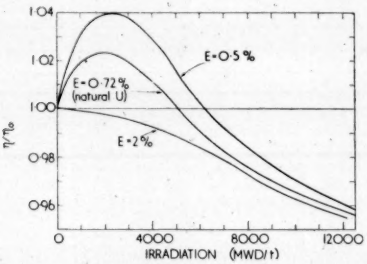

Fig. 3. Reactivity changes on irradiation as a function of enrichment (stated as percentage ^{235}U in ^{235}U–^{238}U mixture) at initial conversion ratio = 1·0 ($\Delta\eta/\eta$ is the change in reactivity)

burnup of *fissile isotope* resulting from a given irradiation decreases as the enrichment increases. Consequently the use of enriched fuel in a once-through cycle increases the waste of fissile isotope. This waste can be avoided by adding the extra fissile isotope to the reactor in the form of separate highly-enriched fuel elements (spiking or seeding) which are reprocessed to recover the unburned fuel. Substantial costs are associated, however, with the reprocessing, refabricating, and inventory of highly-enriched material.

Dunworth[3] has pointed out that a reactor system which initially requires enriched uranium can be operated with natural uranium feed if plutonium is recycled. When this system reaches equilibrium much of the ^{235}U which was initially needed for reactivity purposes has been replaced by an equivalent amount (reactivity-wise) of plutonium, and the actual enrichment $\bar{E}$ at which the equilibrium reactor operates may be less than unity. With some simplifying assumptions, Dunworth arrives at the expression for $\bar{E}$:

$$\bar{E} = E(1 - R_c)(1 + \alpha) \qquad (8)$$

where E is the enrichment required for initial operation of the system and α is that for plutonium, and $\alpha = \Sigma_{capture}/\Sigma_{fission}$ for a given fissile isotope. The system may be fed with natural uranium if $\bar{E} < 1$. His estimates show that for $R_c = 0{\cdot}7$ a reactor with $E = 2$ succeeds in consuming only about 1% of the natural uranium feed, whereas a reactor with $E = 1$ can consume more than 2% of the feed. If, however, $R_c \sim 0{\cdot}85$ can be achieved the consumption for all enrichments between $E = 1$ and $E = 3$ rises to about 4%. Thus, if R_c is high enough the degree of enrichment does not have a large effect. It should be noted, however, that until equilibrium is established, high enrichment can lead to losses of ^{235}U in the burned uranium unless the uranium as well as the plutonium is recycled. Such a loss is avoided, of course, if the initial "enrichment" is provided by plutonium from another reactor rather than by ^{235}U.

Finally, the simple fact must be recognized that other things being equal, the higher the excess of enrichment above unity, the larger the quantity of enriched fissile isotope required to get started. Since the isotope must be supplied by a separation plant or by another reactor it is unlikely to be in unlimited supply.

The foregoing considerations indicate no firm limit to the amount of enrichment that may be used; the advantages of enrichment do not come cheaply, however, and in most cases the tendency will be to use it sparingly.

INCREASE OF CONVERSION RATIO BY ENRICHMENT

For a given set of reactor materials an increase in enrichment makes possible an increase of conversion ratio. Consider an infinitely large reactor which is operating steadily with the equilibrium concentration of ^{135}Xe and ^{149}Sm, and with all control rods out. The conversion ratio R_c is given by

$$R_c = \eta_{25}\epsilon - 1 - R_{xe} - R_{sm} - R_p \qquad (9)$$

where η_{25} is the value of η for pure ^{235}U, R_{xe} is the ratio of absorption by ^{135}Xe to the absorption by ^{235}U, R_{sm} is the corresponding ratio for ^{149}Sm, and R_p is the corresponding ratio for parasitic absorption by moderator, structural material, fuel cladding, etc. R_{sm} is just $Y_{sm}\Sigma_{a25}/\Sigma_{f25} = 0{\cdot}012$, where Y_{sm} is the fission yield of ^{149}Sm. In thermal-neutron fluxes of 10^{14} n cm^{-2} s^{-1} or greater, $R_{xe} = Y_{xe}\Sigma_{a25}/\Sigma_{f25} = 0{\cdot}05$. At lower fluxes R_{xe} is a function of the flux level, having a value 0·028 at $\phi = 10^{13}$ n cm^{-2} s^{-1}.

Thus all the terms of equation (9) except R_p are essentially fixed by the nuclear characteristics of the fission process. To maximize R_c, R_p must be made as low as possible. After all unnecessary materials have been eliminated from the reactor design, further reduction of R_p can be achieved only by increasing the density of ^{235}U in the reactor. At any given enrichment there is a limiting fuel density, beyond which a further increase will so lower the resonance escape probability p that criticality is impossible. Beyond this point R_p can be increased only by further enrichment, and further concentration of the fuel in the reactor.

USE OF ENRICHMENT TO ALLOW HIGHER ABSORPTION CROSS-SECTIONS

Criticality may be attained in thermal reactors having relatively high parasitic absorption cross-sections provided the fuel is enriched to a sufficient degree. The use of enrichment to permit the incorporation of useful materials of medium absorption cross-section (e.g. steel) is a very practical one. The attainment of criticality is not, however, a sufficient accomplishment: one generally desires to have a reasonably high conversion ratio as well, and often considerations of

Table 5. *Characteristics*

Reactor	*Reference No.*	*Nominal power MW*	*Core dia. ft*	*Core height ft*	*Vol. mod. / Vol. fuel*	*Enrichment E*	τ_i	ε
SGR	6	250	15·3	12	24·4	2·52	1·688	1·026
EBWR	19*	20	4	4	3·49	1·70	1·498	1·045 3
D_2O	20	1000	12	12	17·5	1·	1·336	1·029
D_2O						1·7	1·564	1·029

* Engineering design only. Physics data, due to J. Thie, have not been previously published.

conversion rather than those of criticality determine the enrichment to be used.

If the two-group formulation of the reactor equation is used, the equation corresponding to (13) for a finite reactor becomes:

$$R_c = \frac{\eta_{25}\varepsilon}{1+\tau B^2} - (1 + R_{xe} + R_{sm} + R_p)(1 + L^2B^2) - R_{28}L^2B^2$$

$$\simeq \frac{\eta_{25}\varepsilon}{1+\tau B^2} - R_{xe} - R_{sm} - 1 - R_p - \left(1 + \frac{0{\cdot}57}{E}\right)L^2B^2 \quad (10)$$

(since R_{xe}, R_{sm}, R_p, and L^2B^2 are all small)

where $$R_{28} = \Sigma_{28}/\Sigma_{25} \text{ (thermal)}$$

Except for the ratio $\varepsilon/(1 + \tau B^2)$, which will usually be near unity, the first three terms of equation (10) are fixed by the properties of the fission process in a given fissile isotope. The last two terms represent the neutron losses by parasitic absorption and thermal leakage, respectively. They can both be reduced for any given reactor by increasing enrichment, provided, of course, that criticality is maintained by adjusting the lattice arrangement to decrease p. Usually, in a large power reactor, R_p will be the more important term. For such a reactor it can be said, roughly, that if one desires to increase the parasitic absorption (e.g. for engineering reasons), the conversion ratio can be held constant only by a proportionate increase in the ^{235}U content of the reactor. Most of this increase must come from enrichment, although some may result from the closer packing of fuel which accompanies the rearrangement of the lattice to reduce p. An alternate statement is: to hold conversion ratio constant, a fractional increase in the capture cross-section of parasitic materials associated with unit mass of fuel must be compensated by an approximately equal fractional increase in enrichment. The latter statement is particularly appropriate when one considers the addition of parasitic clad or alloying material for the fuel.

Enriched fuel makes possible the use of H_2O as a reactor moderator. The properties of H_2O are given in Table 1, and a typical lattice arrangement is summarized in the last line of Table 3. The slowing down power ($\xi\Sigma_s$) of H_2O is very high, and the maximum-buckling moderator/fuel ratio is consequently quite low. The ratio $(\xi\Sigma_s)/\Sigma_a$ is not quite high enough, however, to make criticality possible with natural uranium. A maximum material buckling of about -2×10^{-4} cm^{-2} has been measured with 1·18 in. diameter natural uranium rods at a water/uranium ratio of 1·36 by volume.[8]

EXAMPLES OF THE USE OF ENRICHMENT

Table 5 summarizes the neutron-physics characteristics of three reactors which may be considered as samples of three reactor types. Since the three reactors represent three different stages of projection into the future, it is not reasonable to compare their over-all performances (the reactor EBWR is a pilot plant, now nearing completion at Argonne National Laboratory; SGR is a proposed intermediate scale power reactor, and the D_2O reactor is a conceptual design of a large-scale power producer). It is instructive, however, to note the details of the neutron economy in the three cases.

SGR is an example of the use of enrichment primarily for the purpose of allowing the introduction of absorbing materials (sodium, zirconium, and stainless steel). This reactor may be compared to the graphite reactor of Table 2, which had a conversion ratio of 0·82 without allowance for fission products. In comparison, SGR loses in conversion ratio about 0·05 to xenon and samarium, and about 0·04 to additional fast leakage. These losses are characteristic of the operating power and of the particular reactor size chosen. If they are subtracted from the conversion ratio of the Table 2 reactor, the remainder is $R_c = 0{\cdot}82 - 0{\cdot}09 = 0{\cdot}73$, just slightly more than that of SGR. Thus, in the SGR design, the conversion ratio has been preserved as absorber has been added by increasing E and lowering p. From the detailed neutron balance presented in reference 6 it is apparent that the absorbers added to the SGR have increased the absorption per moderator atom by about a factor of 3·6 over the "clean" case of Table 2. In SGR, however, the moderate/fuel ratio has been decreased to 24, and the ratio of absorption per *fuel* atom has therefore been increased only by the factor 3·6 (24/44) = 2·0. This is roughly the factor by which E would be expected to increase to preserve the conversion ratio. This estimate, which ignores changes in disadvantage factors, is somewhat lower than the value of 2·5 quoted for SGR on the basis of detailed calculations (Table 5).

The EBWR reactor is, of course, an example of the use of enrichment to make possible a reactor moderated by ordinary water. It is discussed in more detail later.

The 1000 MW D_2O boiling reactor was proposed as a once-through-natural-uranium plant with a target burnup of 10 000 MWD/ton. W. B. Lewis has estimated, however, that on the basis of the quoted conversion ratio the reactor with plutonium recycling could irradiate the natural-uranium fuel to about 32 000 MWD/ton.[7] He has suggested further that the burnup might be extended to 45 000 MWD/ton if the enrichment is sufficiently increased to lower p to 0·70. The last line of Table 5 shows roughly the reactor characteristics corresponding to such a change (estimated assuming ϵ and disadvantage factors do not change). This modification illustrates the use of enrichment to increase conversion ratio. The recycling of plutonium is, of course, far superior to once-through operation from the standpoint of conservation of uranium resources. Whether it is a profitable mode of operation at any given stage in the development of a nuclear power industry depends upon the economics of fuel-element fabrication from recycled material.

of three power reactors

p	f	f_{mod}	f_p	k	τ cm^2	$\frac{B^2}{10^4}$ cm^{-2}	k_{eff}	R_c
0·7564	0·8551	0·0283	0·123	1·120	435	2·4	1·02	0·72
0·8193	0·848	0·1064	0·0453	1·089	57	15·08	1·00	0·74
0·8364	0·932	0·01	0·06	1·0717	184	1·74	1·023	0·901
0·700	0·951			1·0717				0·94

f_{mod} = fraction of thermal absorptions by moderator alone.
f_p = fraction of thermal absorptions by parasitic absorbers (Xe, Sm, and structural materials).

SPECIAL CHARACTERISTICS OF H_2O-MODERATED REACTORS

The relatively large fast-fission effect characteristic of closely-packed lattices in H_2O is well known. The enhancement of the effect arises from the fact that fission neutrons have a relatively high probability of travelling from one fuel element to another in the lattice before experiencing a slowing-down collision. Basically this situation stems from the combination of the very high slowing-down power of water in the resonance region (therefore a tight lattice) with a reasonably long mean free path for neutrons of fission energy. Values of ϵ of 1·105 and 1·061 have been measured for water/uranium volume ratios of 1 and 2, respectively.[8] Since a given percentage change in ϵ produces roughly twice the percentage change in R_c, the fast fission effect is quite important to conversion ratio. In addition, it represents the direct fissioning of some ^{238}U.

For engineering reasons, H_2O is strongly preferred as the reflector material for H_2O-moderated reactors. Since it gives only a modest reflector saving, large H_2O cores of uniform composition have a relatively high maximum/average power density ratio. The short migration length in the reactor makes it possible, however, to employ power flattening by space-variable composition without incurring a large penalty in increased neutron leakage. Fig. 4 shows the calculated power flattening in EBWR due to non-uniform fuel burn-up if the reactor is initially loaded uniformly and the fuel element positions are unchanged until an average burn-up of 3000 MWD/ton.

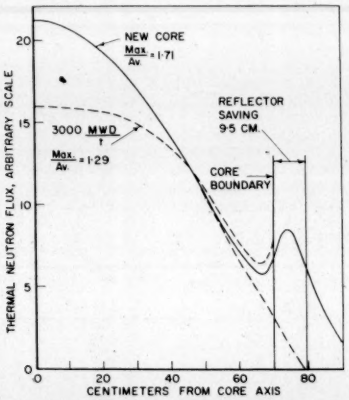

Fig. 4. Radial thermal flux distributions in reactor EBWR at startup and after approximately 3000 MWD/t irradiation of fuel (no steam in reactor). (Calculation by O. A. Schulze)

Because of the rather large absorption cross-section of H_2O itself, it is necessary to make the total absorption cross-section of a water-moderated core quite high in order to achieve criticality. The macroscopic absorption cross-section in the core of EBWR at room temperature is about 0·07 cm^{-1}. This "blackness" of the core leads to difficulties in achieving a sufficiently large amount of variable reactivity for control. A total control rod worth of about 15% k_{eff} was desired for EBWR. If it is assumed that thermally-black control rods are to be used, the rod requirements can be estimated very roughly as follows:

$$\text{Required} \quad \frac{\Delta k}{k} = \frac{\Delta f}{f} = \frac{\Delta \Sigma_a}{\Sigma_a} = 0{\cdot}15$$

where Σ_a is the value after the absorber has been added. Thus the control rods must increase the absorption by an effective cross-section of $\Sigma_{a\,eff} = 0{\cdot}012\ cm^{-1}$. If the reactor core is divided into a number of cells of square cross-section,

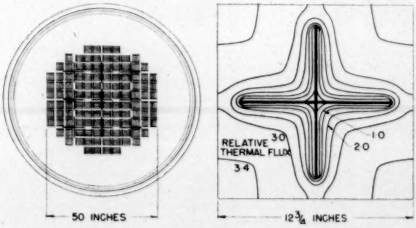

Fig. 5. Control rod pattern in reactor EBWR and thermal flux contours around a single rod in an infinite lattice of rods. (Diffusion theory calculation by O. A. Schulze)

each defined by a set of four thermally black cruciform control rods (Fig. 5), the neutron current density into the black cell boundary is, roughly,

$$J = \frac{\lambda_{tr}}{3}\frac{\phi}{L},$$

where ϕ is the flux at the centre of the cell. Since L is small compared to a, the length of side of the cell, the control requirement is, approximately:

$$4a\left(\frac{\lambda_{tr}}{3}\frac{\phi}{L}\right) = 0{\cdot}012\phi a^2;\ a = \frac{4}{0{\cdot}012}\cdot\frac{\lambda_{tr}}{3L} = \frac{4}{0{\cdot}012}\cdot L\Sigma_a\ \text{(core)}$$

In EBWR, $L \simeq 1{\cdot}8$ cm, giving $a = 42$ cm. Detailed calculations, including the effects of gaps at the rod edges, and the transport correction at the cell boundary, give $a = 32{\cdot}4$ cm

($= 12\frac{4}{4}$ in.) (Fig. 5). This relatively high density of control rods in the reactor may be considered to be due to the circumstance that, so far as thermal neutrons are concerned, only those portions of the core quite near the control rod feel the effect of the rod. In Fig. 5 is included a calculated thermal flux contour around one of the control rods which illustrates this point.

A related effect of the short diffusion length is the so-called "water-hole effect." If the normal lattice structure of the core is interrupted by an extra volume of H_2O which does not contain other strongly absorbing material, most of the neutrons slowed down in that volume of water will be absorbed in the immediately adjacent fuel elements. The result, of course, is a local peak in the fission power. Fig. 6 shows the

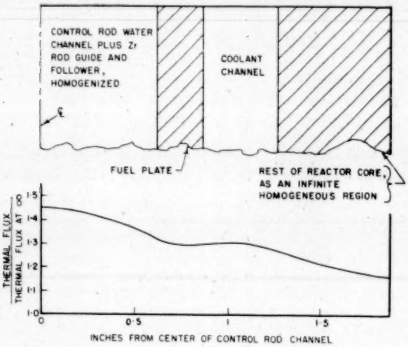

Fig. 6. Thermal flux peaking in a control rod channel when the control rod is withdrawn. (Diffusion theory calculation by O. A. Schulze)

calculated peaking of thermal-neutron flux in one of the EBWR control rod channels when the rod is withdrawn, and the accompanying extra flux in the adjacent fuel plate.

SPECIAL CHARACTERISTICS OF BOILING REACTORS

Up to this point two design requirements have been applied to the physics of power reactors: that of criticality and that of an acceptable conversion ratio. The boiling reactor is subject to a further requirement: that of a proper value for the steam coefficient of reactivity. When steam displaces a finite amount of water from the reactor core the reactivity will inevitably change. If the change is in the positive direction the reactor is autocatalytic, and will "run away." If the change is in the negative direction the rate of formation of steam in the core will regulate itself at some value consistent with the amount of reactivity available and the rate of flow of steam out of the core. Experiments have shown that such self-regulation is stable in a natural-circulation reactor only so long as the total steam content of the reactor represents a negative reactivity of not more than 2% or 3% k_{eff}. The steam coefficient of reactivity C_s is defined as:

$$C_s = \frac{k_{eff} \text{ with steam in core} - k_{eff} \text{ without steam in core}}{\text{fraction of coolant volume occupied by steam}}$$

C_s may be adjusted by adjustment of p, f, and leakage. Untermyer has discussed[9] the conditions for negative C_s. However, for high power density in natural-circulation reactors C_s must be made small as well as negative. The difficulty in achieving both requirements over the range of operating conditions of the reactor results from the fact that C_s is not constant, but tends to become more negative as the molecular density of H_2O in the reactor decreases, whether the decrease be due to increased steam content or higher temperature.

Table 6 illustrates the situation by giving the components of the reactivity change with steam content, both at room temperature and at operating temperature. At 20° C (68° F) the decrease in p as water is displaced by steam is more than compensated by the increase in f, and the net change in k is positive. The change in k_{eff}, however, is very slightly negative because of the increase of M^2, and hence of leakage. At 250° C (486° F) the reactor is sufficiently undermoderated that the effect of water removal is larger on p than on f, and the net change in k is negative. When the effect of higher leakage is added in, the net value for C_s is still acceptable, but rather higher than is desired.

It is evident that leakage is not necessary for the attainment of a negative steam coefficient. If the reactor were larger, one would simply decrease p slightly below the initial value in Table 4. It is evident also that the moderator/fuel ratios which give proper values of C_s will be in the range of interest from the standpoint of conversion ratio.

In D_2O-cooled boiling reactors the balancing of the effects of water displacement on p and f does not occur, and the dependence of reactivity on steam content is much more nearly linear. An extra degree of freedom exists in adjusting C_s in that p can be varied by varying the clumping of fuel elements, as well as by varying moderator/fuel ratio.

A further characteristic of the boiling reactor is the variation in steam content, and hence in moderator/fuel ratio, along the length of the reactor. Fig. 7 shows the calculated axial neutron flux distributions resulting from an estimated distribution of steam in the EBWR. The distortion of the flux distribution is not serious in this case. It will become more pronounced in a longer reactor, for which the axial buckling will be lower and the reactivity, therefore, less

Table 6. *Effect of steam in*

Temperature / *F*	*Vol. steam* / *Vol. water*	τ_s	τ	p	f	k
68	0	1·535	1·037	0·870	0·787	1·090
68	0·1	1·535	1·039	0·859	0·800	1·096
Fractional change / Steam fraction			+0·019	−0·128	+0·162	+0·055
486	0	1·535	1·042	0·840	0·813	1·093
486	0·15	1·535	1·045	0·819	0·828	1·089
Fractional change / Steam fraction			+0·019	−0·17	+0·12	−0·024

sensitive to the increases in buckling which accompany departures from a symmetrical distribution.

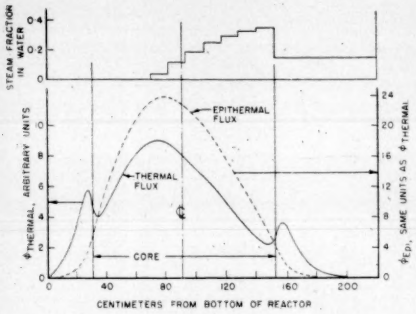

Fig. 7. Axial flux distributions in reactor EBWR with assumed steam distribution. (Calculation by O. A. Schulze)

USE OF THORIUM–^{233}U FUEL

The thorium–^{233}U fuel cycle has long been regarded as a most attractive one because of the high value ($2{\cdot}31 \pm 0{\cdot}03$) of η for the thermal fission of ^{233}U. Since η for ^{235}U under the same conditions is about 2·08, the ^{233}U–Th system has potentially a considerably higher conversion ratio than the ^{235}U–^{238}U system. Some of this advantage is offset by the low value of the fast-fission effect in thorium, but if an advantage of 0·06 is allowed to ^{238}U for this effect, the Th–^{233}U system still has a net potential ratio which is higher by about $2{\cdot}31 - 2{\cdot}08 - 0{\cdot}06 = 0{\cdot}17$.

The higher value of η does not mean, however, that the reactor designer will strive any the less for a high value of R_c. On the contrary, the fact that there is no natural supply of the fissile isotope in the cycle places a large premium on the attainment of a total conversion ratio (including processing losses) of at least 1·0. Even after this goal is attained, the desire to produce a net output of ^{233}U for the start of other reactors will be a further incentive to increase of R_c. Indeed, the most important problem in the large-scale use of the ^{233}U–Th cycle is probably that of obtaining sufficient enriched fissile isotope to start a large number of reactors.

The design principles for the ^{233}U–Th reactor are much the same as those previously considered. There is, of course, no "natural" ratio of fissile to fertile material. The ratios used in reactors will usually be considerably higher than the fissile/fertile ratios in the ^{235}U–^{238}U cycle simply because of the high thermal absorption cross-section of thorium. Thus, a reactor having $p = 0{\cdot}85$ and $k = 1{\cdot}07$ would require, for criticality, an atomic ratio ^{233}U/^{232}Th = 0·0143 even if no other absorbers were present. A high value of this ratio does not necessarily indicate a high ^{233}U inventory, at least from the neutron-physics standpoint, since the quantity of thorium in the reactor need be only that amount which will absorb the desired number of neutrons for conversion. If thorium is the only material used as fuel diluent, however, considerations of heat removal and radiation damage may also have a bearing on the quantity used.

Although the resonance integral for thorium at infinite dilution is less than $\frac{1}{3}$ that of ^{238}U, the difference is not great in the region of moderator/fuel ratio which has interest for solid fuel power reactors.[10] The difference that does exist, however, along with the lower atomic density of thorium, will no doubt lead to reactor designs with appreciably lower volume ratios of moderator/fuel than in the ^{235}U–^{238}U case. Starr[6] gives comparative designs with ^{238}U and ^{232}Th as fertile isotope which illustrate this point.

THE FAST REACTOR

The fast reactor occupies an important place in the long-term future of nuclear power because of its excellent breeding possibilities. Although values of η appropriate to the neutron energy spectrum of the fast reactor have not been measured, there is much experimental evidence, including actual breeding ratio measurements on EBR–I[11] and ZEPHYR[12] that breeding ratios substantially above 1 can be attained. The fast breeder reactor is at present the only known possibility for utilizing a major fraction of the energy potentially available in ^{238}U.

Although η has not been measured at high energies, α measurements have been made (α = ratio of radiative capture cross-section of the fissile isotope to fission cross-section). Since it is expected, and there is some experimental evidence,[13] that ν, the number of neutrons emitted per fission, will increase slightly with neutron energy, it is a conservative procedure to use known or estimated values of α with the thermal-fission value of ν as an approximation to η for fast neutron fission.

The following expression has been used for the breeding (or conversion) ratio, R_c, in terms of ν and α:

$$R_c = \frac{\nu - 1 - \alpha - A - \Lambda + F(\nu' - 1)}{1 + \alpha}$$

where

A = number of neutrons captured by parasitic absorbers
Λ = number of neutrons leaking from the reactor (core plus blanket)
} per fission of primary fissile isotope

F = number of ^{238}U atoms fissioned
ν refers to the fissile isotope (^{235}U or ^{239}Pu) and ν' refers to the fertile isotope (^{238}U).

EBWR calculated by J. A. Thie

τ	L^2	*R.S.*	B_g^2 10^4 cm^{-2}	M^2	$1 + M^2B_g^2$	k_{eff}
34·75	3·02	7·02	16·66	37·77	1·0630	1·0252
39·74	3·32	7·54	16·42	43·06	1·0707	1·0229
					+0·071	−0·022
47·09	6·53	9·50	15·55	53·62	1·0834	1·0082
56·96	7·49	10·62	15·08	64·45	1·0972	0·992
					+0·084	−0·107

Zinn[14] has estimated on the basis of thermal values of ν, that values of R_c in the ranges 1·2 and 1·7 can be attained with ^{235}U and ^{239}Pu, respectively.

The high values of R_c attainable in a fast reactor result from the following circumstances:

(*a*) The value of α is low for neutrons of very high energy.

(*b*) The value of F may be made large in an unmoderated neutron energy spectrum. F is estimated to be in the range of 0·3 for a large fast-breeder reactor.

(*c*) The ratio of fuel to other materials in a fast reactor is so high that A is usually quite small.

Since α increases with decreasing neutron energy, and the fission threshold for ^{238}U is about 1 MeV, the characteristics (*a*) and (*b*) above are exploited most effectively if the neutron energy spectrum is kept as high as possible. Consequently, in the fast breeder reactor, materials with high moderating power ($\xi\Sigma_s$) are excluded, and it is desirable further to keep the inelastic scattering as low as possible. The typical composition for the fast reactor, then, consists of a fissile isotope (^{235}U or ^{239}Pu), a fertile isotope (^{238}U), a non-moderating coolant (sodium or an alloy of sodium and potassium), and structural and containing materials, usually metals of medium atomic weight (e.g. stainless steel). Alternatively, the fissile-fertile combination could be ^{233}U–^{232}Th.

A rough idea of how these materials are combined in the reactor may be had from the following considerations. For neutrons in the energy range 0·1–1·0 MeV, the scattering mean free path in pure ^{235}U is about 2 cm and the absorption mean free path is about 10 cm. If one writes for this medium and energy range an effective diffusion area, L^2:

$$L^2 \simeq \frac{\lambda_s\lambda_a}{3} \simeq \frac{(2)(10)}{3} \simeq 7\ \text{cm}^2$$

If this material is considered the starting point for a fast reactor, one might consider first diluting the material by a coolant, say in the volume ratio 1/1. If the coolant is sodium, which is practically transparent to neutrons in the energy range of interest, L^2 is increased to:

$$L^2 = (7)(2)^2 = 28\ \text{cm}^2$$

One might then consider maintaining this ratio of coolant to solid material, but diluting the ^{235}U further by replacing some of it by ^{238}U and structural materials. Such materials are unlikely to have a scattering mean free path less than that of uranium. Furthermore, the diluting materials cannot be allowed to absorb more than about half the neutrons if a chain reaction is to be maintained. Consequently, if the dilution (= volume total solid material/volume ^{235}U) is denoted by D, one can say, very roughly:

$$L^2 \text{ lies between } 28D \text{ and } 28D/2\ \text{cm}^2$$

If significant conversion is to be done in the reactor core, k will not be extremely high, and the reactor dimensions must be several diffusion lengths in magnitude for criticality. Hence D cannot be a very large number for a reactor of reasonable size. Values for proposed high-power breeder reactors lie in the range 5 to 10.

Because of the high concentration of fissile isotope in the fast reactor, the parasitic absorption of structural materials (and fission products) is not so important as in the case of thermal reactors.

Table 7 lists some results of danger coefficient measurements from a ZPR–III reactor assembly having the approximate composition of EBR–II. The desirable structural materials have effective absorption cross-sections of the order

Table 7. *Danger coefficient measurements in ZPR-III*
(Preliminary Data)

Measurements were made with a cubic sample of 8 in^3, at the centre of the assembly. Results are quoted in millibarns effective absorption cross-section, normalized to $\sigma = +3{\cdot}65$ barns for ^{239}Pu. The positive sign indicates a gain in reactivity; the negative sign indicates a loss.

Material	σ_{eff}	*Material*	σ_{eff}
C	+ 30	Ru	−137
Si	0	Pd	−258
S	− 30	Ag	−220
Ti	0	Cd	−100
V	+ 12	Sn	− 37
Cr	− 14	Sb	−130
Mn	+ 6	I	−200
Fe	− 13	Ce	− 35
Co	− 3	Hf	−179
Ni	− 20	Ta	−207
Cu	− 20	W	−100
Zn	− 21	Hg	− 63
As	− 73	Pb	− 11
Se	− 16	Bi	− 8
Zr	− 21	Th	−137
Cb	− 82	U^{238}	+ 18
Mo	− 48		

1/100 that of ^{239}Pu. Although the effective cross-sections include the effect of inelastic scattering, they are indicative of the general magnitude of absorption. In the fast reactor the enrichment and the fuel concentration are so high that these structural materials are used only in volumes comparable with the volume of fissile material (cf. Fig. 8). Consequently, they do not represent a serious waste of neutrons.

Because the materials suitable for use in fast reactors do not have widely differing properties in the fast-neutron spectrum, it seems improbable that solid-fuel fast reactor designs will exhibit a diversity of neutron-physics characteristics comparable to that in the thermal-reactor field. At least the designs which are currently under consideration are qualitatively quite similar, and it is possible to illustrate their general characteristics by the results of calculations on a few idealized cases. Fig. 8 gives the sizes and compositions of

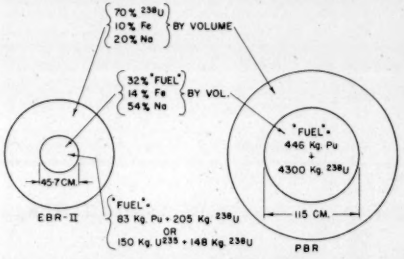

Fig. 8. Compositions of spherical reactors whose characteristics are given in Table 8

two of several spherical reactors calculated by Okrent, Avery, and Hummel.[15] The smaller reactor has the approximate composition of EBR–II,[16] while the larger one is repre-

sentative of a large fast breeder power plant. Some of the calculated characteristics of these two reactors are given in Table 8. Note the high proportion of ^{238}U fissions and the low parasitic absorption.

Table 8. *Calculated characteristics of spherical reactors approximating EBR-II and PBR*

(Reproduced from Reference 15)

	EBR-II (^{235}U)	*EBR-II* (*Pu*)	*PBR* (*Pu*)
Median fission energy in core (MeV)	—	0·49	0·36
k	1·90	2·18	1·48
$M = \sqrt{\frac{(k-1)}{B^2}}$ (cm)	11·7	13·4	16·7
$\frac{^{238}U \text{ fissions}}{\text{fissions in } ^{235}U \text{ or Pu}}$	0·265	0·362	0·371
Internal breeding ratio	0·090	0·187	0·855
Total breeding ratio	1·31	1·89	1·87
Parasitic absorption in core	0·6%	0·7%	1·2%
Neutron lifetime	—	$0{\cdot}8\times10^{-7}$	$1{\cdot}7\times10^{-7}$

As the reactor is made larger and more dilute the effective energy of the neutrons in the reactor core decreases because of the relative increase in inelastic and elastic scattering. Fig. 9 gives calculated core spectra for the two reactors of Fig. 8, as well as for EBR–I,[11] a reactor having a core of highly enriched ^{235}U. The EBR–I reactor has been used for measurements pertaining to the larger reactors despite its faster core spectrum. In the blanket of natural uranium surrounding the core (Fig. 10) the spectrum is degraded by inelastic and elastic scattering, and hence varies with the radial distance from the core. The calculations indicate that it is roughly equivalent to that of the large power breeder in the region marked on Fig. 14. Values of α for ^{235}U and ^{239}Pu, measured radiochemically at various points in the reactor are plotted in the figure.

The wide range of neutron energies over which the important processes occur in a fast reactor makes necessary a large body of detailed cross-section data for the precise

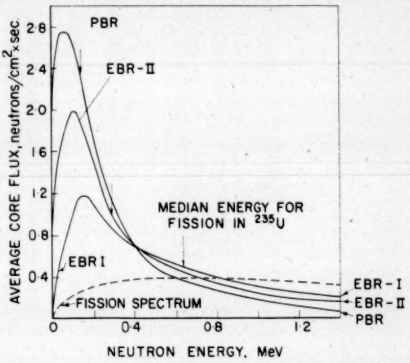

Fig. 9. Calculated neutron spectra in the cores of three fast reactors.

calculation of fast reactor characteristics. Since a complete set of such data does not yet exist, the critical experiment occupies an especially important place in the study and design of fast reactors. Zero Power Reactor III (ZPR–III) has been constructed at the Idaho Division of Argonne National Laboratory for the experimental design of EBR–II, and for general studies on fast reacting systems.[17] Fig. 11 shows

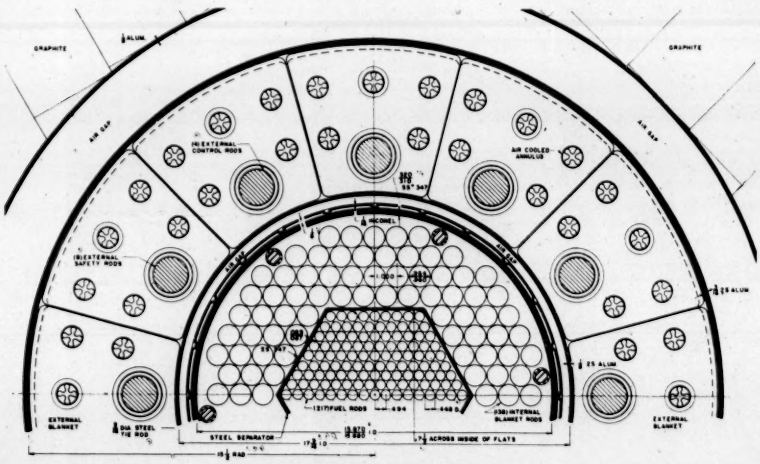

Fig. 10. Partial cross-section of EBR–I core

the fission distributions taken with ^{235}U and natural-uranium fission counters in an assembly having approximately EBR–II composition. The relatively faster drop-off of the natural-uranium counting rate in and near the blanket demonstrates the softening of the neutron spectrum in those regions.

the internal (core) breeding ratio is not high; the large total breeding ratio is achieved by catching leakage neutrons in the blanket. Recovery of bred material from a fast-reactor blanket is almost certainly a profitable operation, whereas the corresponding operation on a thermal reactor may be

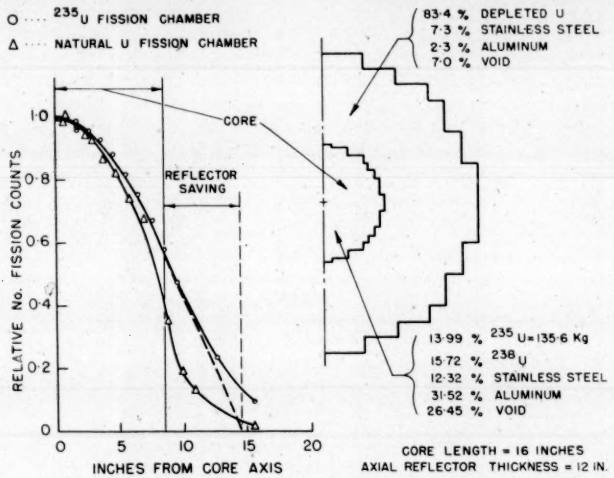

Fig. 11. Fission count distributions in ZPR–III. Preliminary data from Argonne ZPR–III group. (Reference 23)

The danger coefficients of Table 7 were taken at the centre of an assembly of the same core composition and slightly reduced blanket density. It should be pointed out that since these coefficients give the net effect of absorption and scattering they are characteristic not only of the neutron energy spectrum, but to some extent of the leakage from the reactor as well.

The ZPR–III is constructed of plates of ^{235}U, ^{238}U, stainless steel, and reduced-density aluminium (to simulate the sodium of EBR–II). Since the plates are only $\frac{1}{8}$ in. thick a relatively close approach to homogeneity of composition can be made. Nevertheless, experiments were made to determine whether there were any important effects of neutron streaming along the aluminium plates or of material lumping. No important streaming effects were observed, and the lumping effects were shown to be small in the assembly structure normally used. When the lumping was purposely exaggerated by grouping similar plates, the reactivity was shown to be affected markedly by the lumping, and to some extent by the position of ^{238}U relative to ^{235}U, the reactivity being highest when the two materials were adjacent. The fission rate measurements through a typical section of the structure (Fig. 12) show the enhancement of the ^{238}U fission rate near the ^{235}U, where the density of uncollided fission neutrons is highest. Such effects are consistent with the results of multi-group calculations using the S_4 transport theory approximation.

As a final remark on the fast reactor system it should be pointed out (cf. Table 8) that except in very large fast reactors

marginal economically. There are two primary reasons for this situation:

(*a*) The concentration of fissile isotope builds up much more rapidly in the fast-reactor blanket because of the higher power density and the larger number of neutrons available for conversion.

(*b*) Even with a low internal conversion ratio the fast

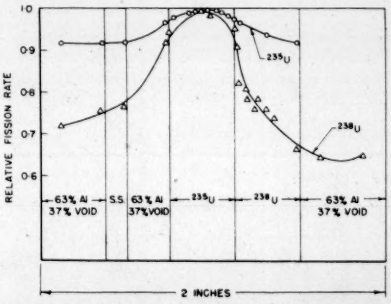

Fig. 12. Local fission distributions in a lumped loading of ZPR–III. Preliminary data from Argonne ZPR–III group. (Reference 23)

reactor does not lose reactivity rapidly with fuel burn-up. Because of the high ratio of fissile to fertile material, a given percentage burn-up of material in the fuel element does not represent a large percentage loss of fissile atoms.

SAFETY OF FAST REACTORS

Although it is by no means necessarily true that the fast reactor is less safe than the typical thermal power reactor, it can be said that the fast reactor possesses, to a unique degree, certain reactor characteristics which add to the difficulty of an absolute proof of reactor safety.

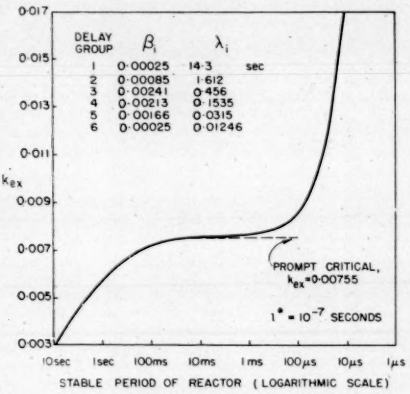

Fig. 13. The relation between excess reactivity and stable period for a reactor of neutron lifetime 10^{-7} s

The short neutron lifetime, of the order 10^{-7} s (Table 8) has been frequently cited as one of the more difficult characteristics of the fast reactor. It results in a very steep slope of the curve of reactor period as a function of excess reactivity in the neighbourhood of prompt criticality (Fig. 13). Thus, the chances of attaining a very short period in a hypothetical reactivity accident are greater for a fast reactor than for a thermal reactor, for which the lifetime will lie in the range 10^{-5}–10^{-3} s. Theoretical treatments of such hypothetical accidents generally predict that the shorter reactor periods will lead to larger total energy releases in the accident, and greater destructiveness. The situation is particularly dangerous if there is any possibility of a positive temperature coefficient of reactivity, for such a coefficient need add only a small increment of reactivity to shorten the period from tenths of seconds to tenths of milliseconds if the reactivity is already in the neighbourhood of prompt criticality.

Since theory predicts the possibility of a positive temperature coefficient in highly enriched systems because of Doppler broadening of fission resonances,[18] an experimental search is being made for such an effect in critical systems of various compositions in ZPR–III. Measurements by W. Y. Kato, on a system having the approximate EBR–II composition, have so far had a sensitivity limit about a factor of four above the theoretically predicted effect, and as might be expected, have shown no effect. The technique used is that originally used in a reactor of lower neutron energy spectrum by R. T. Frost of Knolls Atomic Power Laboratory, and consists of oscillating the temperature of a ^{235}U helix in the critical assembly and observing the magnitude of any power fluctuations which result. It is anticipated that increases in the size of the heated sample with respect to the size of the critical assembly, increases in operating power of the assembly, and improvements of instrumentation will make possible the detection of any effect of practically significant magnitude.

Fortunately, the Doppler broadening of absorption resonances in ^{238}U is expected to over-ride any positive reactivity component in reactors which use considerably greater quantities of ^{238}U than ^{235}U, and use the two isotopes

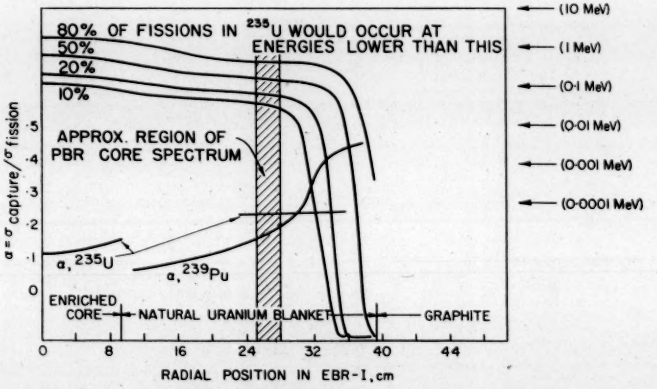

Fig. 14. Values of ν measured in EBR–II (Reference 15). Radiochemical measurements of α by Argonne Chemical Engineering Division (Reference 22)

in intimate mixture. Thus, for large fast breeders the Doppler effect may increase safety, as it does in typical thermal reactors.

Fully as important as the short neutron lifetime is the circumstance that practical fast power reactors typically consist of several potential critical masses, which collectively are just critical in the reactor because they are diluted by the coolant. A result of this situation is that one can imagine numerous configurations of core material which would be highly supercritical if the normally solid material were allowed to arrange itself in different patterns following an accident which initially caused it to melt. A second effect is that the reactor normally has a relatively high compaction coefficient of reactivity. This coefficient C_c for a bare reactor is given by

$$\frac{\Delta k_{eff}}{k_{eff}} = -\frac{M^2B^2}{1+M^2B^2}\left(\frac{\Delta M^2}{M^2}+\frac{\Delta B^2}{B^2}\right)$$

$$= -\frac{M^2B^2}{1+M^2B^2}\left(6\frac{\Delta R}{R}-2\frac{\Delta R}{R}\right)$$

$$C_c = \frac{\Delta k_{eff}/k_{eff}}{\Delta R/R} \simeq -\frac{4M^2B^2}{1+M^2B^2} \simeq -4\frac{k-1}{k}$$

where R is the reactor radius. The effect of a reflector which is assumed not to participate in the compaction will be to reduce the coefficient by some factor, of the order 2, depending on the core and reflector size and composition.

It is evident that for small reactors small changes in effective core radius will produce rather large reactivity effects, and it may be inferred that even small core distortions such as those that might result from thermal expansions in regions of sharp temperature gradient might produce significant reactivity effects.

The EBR–I reactor has exhibited a "prompt" positive temperature coefficient, of magnitude about 1/3 that of the "slow" (isothermal) negative coefficient of $-1{\cdot}5$ inhours per degree C. Although this positive coefficient does not present any difficulty in the normal operation of the reactor, an experimental programme is underway to identify its source. Possibilities are the Doppler effect and transient thermal distortions of the reactor core. It was during an experimental power excursion in this programme that the reactor core was overheated and damaged in November, 1955[21].

In current fast reactor projects the peculiar hazards of fast reactors are being eliminated by appropriate specific design features. Since the necessity for such ingenious design features involves penalties in performance and cost, it may be predicted that the basic physics of fast reactor safety will be an important field of investigation in the immediate future.

APPENDIX

Symbols and definitions

E = Enrichment = $\dfrac{\text{fraction } ^{235}\text{U in } ^{235}\text{U}\text{–}^{238}\text{U mixture}}{\text{fraction } ^{235}\text{U in natural U}}$

f = Thermal utilization = $\dfrac{\text{thermal neutron absorption in fuel}}{\text{total thermal neutron absorption}}$

k = Infinite multiplication constant (see equation 1)

k_{eff} = Effective multiplication constant for a finite reactor = $\dfrac{\text{number of neutrons in generation } (n+1)}{\text{number of neutrons in generation } n}$

L^2 = Diffusion area = $\frac{1}{6}$ the mean square distance travelled by a thermal neutron in a medium during its lifetime (cm^2).

M^2 = Migration area = $L^2 + \tau$ (cm^2).

p = Resonance escape probability. In an infinite medium of the same composition as the reactor, the number of neutrons reaching thermal energy per neutron slowing down past the fast-fission threshold of the fertile material in the fuel.

R_c = Conversion (or breeding) ratio. The number of atoms of new fissile material formed per atom of fissile material destroyed.

$R.S.$ = Reflector saving = amount by which the reflector increases the effective size of the reactor core.

α = For a fissile isotope, the ratio of the cross-section for radiative capture of neutrons to the fission cross-section.

ϵ = The fast fission factor: The number of neutrons slowing down past the fission threshold of the fertile material per neutron produced by thermal fission (medium assumed infinite).

η = The regeneration factor: The number of fission neutrons produced per neutron absorbed in fuel.

λ = Neutron mean free path ($= 1/\Sigma$) (cm).

λ_{tr} = Transport mean free path (cm).

ν = Average number of neutrons emitted per fission by a given isotope.

ξ = Average logarithmic energy loss of a neutron per collision with a nucleus of the medium.

Σ = Macroscopic neutron cross-section (cm^2 per cm^3 of material, or cm^{-1}).

Σ_a = Macroscopic absorption cross-section.

τ = The Fermi age for neutrons in the medium = $\frac{1}{6}$ the mean square distance travelled by a neutron between the time of its birth by fission and the time it becomes thermalized.

Inhour = A unit of reactivity $\simeq 2{\cdot}5 \times 10^{-5}\, k_{eff}$.

MWD/ton = A unit of irradiation for fuel = 1 megawatt day of fission energy produced per ton of fuel.

Fuel is used to designate the mixture of fertile and fissile isotopes in the reactor.

EBR–I = Experimental Breeder Reactor I—A1 MW fast reactor fuelled with ^{235}U and cooled with sodium–potassium alloy, at Argonne's Idaho Division (reference 11).

EBR–II = A larger, sodium-cooled fast breeder now under design (reference 16).

EBWR = Experimental Boiling Water Reactor. A 20 MW boiling H_2O reactor under construction at Argonne National Laboratory (reference 19).

SGR = Sodium Graphite Reactor. A proposed reactor moderated by graphite and cooled by sodium (reference 6).

Zephyr = A low-power, uncooled fast reactor using plutonium as fuel, at Harwell (reference 12).

ZPR–III = Zero Power Reactor–III. A critical assembly facility for fast reactor measurements at Argonne's Idaho Division (reference 17).

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DISCUSSION ON SESSION 2

Dr. G. C. Laurence: The plans for nuclear electric power in Canada are somewhat different from the plans in the United Kingdom, not because there are important differences of opinion on technical questions but owing to economic differences. Over 90% of the electricity consumed in Canada is produced from water power at a cost of about 4 mills/kWh. It will be a long time before electric energy from nuclear power can compete with these low costs. However, in some parts of the country like Southern Ontario new power requirements will have to be met through the use of thermal stations. It will cost at least 6 mills/kWh to produce electricity from the burning of coal; and I foresee the cost of producing electricity in this region will have increased considerably for this reason by 1965. We expect that by then nuclear electric power will become a cheaper alternative to the use of coal fired stations for new power needs.

The second important factor is that Canada is a uranium producer. Its known ore reserves have been increasing at a rate that gives confidence that there will be abundant nuclear fuel for Canada's own requirements and a surplus for export. For this reason we are interested in the development of reactors that use uranium or its derivatives for fuel, rather than fuel derived from thorium, and there is very little interest in the development of breeder reactors.

Thirdly, it is more appropriate to the scale of Canada's industrial resources and technical man power to concentrate available effort on only one type of reactor, and that must be the one which seems to be most suitable for Canadian requirements. The first question is the kind of fuel to use. Concentrated ^{235}U is an expensive fuel, at a price of $25/g it would cost over 2s. 6d. to produce a million B.t.u. of heat energy, even if all the latent energy in ^{235}U were recovered. This is no cheaper than the use of coal in Canada. By the use of natural uranium, the cost of the extraction of ^{235}U is avoided. Most of the energy can be obtained from the ^{235}U content and several times as much energy from the ^{238}U content, through its conversion to plutonium in the reactor.

Two conditions must be fulfilled to obtain this high energy yield from natural uranium. First, the fuel must not fail mechanically from radiation damage. Metal rods of natural uranium tend to disintegrate after yielding about 3000 MWD/t, but it has been demonstrated recently both in a number of American laboratories and at Chalk River that uranium oxide will withstand the large energy release required without mechanical failure. The second requirement is the fulfilment of the critical condition of reactivity. As energy is extracted from natural uranium, its reactivity decreases because ^{235}U is being replaced by the less reactive plutonium and because fission products are accumulating. To obtain a very high energy release it is necessary therefore to enrich the reactor by the addition of some more concentrated fuel; ^{235}U may be used for this purpose. It is cheaper to use natural uranium with some concentrated ^{235}U than it is to use natural uranium alone or to use concentrated ^{235}U alone. An alternative to the use of ^{235}U for enriching the reactor is the use of plutonium. The plutonium would be recovered from old rods of natural uranium after they had been removed from the reactor. The recovered plutonium would be placed in the reactor again with the next loading of fuel. This is profitable when it can be done on a sufficiently large scale to justify the capital investment in the plutonium extraction plant. With a power output of over a million horse power it is expected that the fuel cost using uranium with the recycling of plutonium residues should not exceed 1½ mills/kWh.

The next question is the choice of the moderator and the choice of the fluid used for cooling the reactor. Heavy water is a very expensive liquid; however, it is a very much better moderator than graphite and smaller quantities are required. It is much less wasteful of neutrons than natural water or sodium if used as a coolant. Under conditions in Canada the use of heavy water appears to be most economical both for moderating and for cooling the reactor.

As the first step in the Canadian reactor development programme a small plant to be known as NPD is now being designed and is planned for construction near Chalk River and to be completed in 1959. The fuel will be pellets of uranium oxide enclosed in zirconium tubes. The heavy water for cooling will flow through a zirconium tube 3½ in. in diameter surrounding the uranium oxide rods. This will be placed inside another zirconium tube of slightly larger diameter, thereby separating the heavy water for cooling from the heavy water for moderating by a small annular gap filled with helium gas. This gap will retard the outward flow of heat from the coolant to the moderator and so make it possible to maintain the moderator at a much lower temperature.

There will be no mechanical control rods in this reactor; control of power is effected by controlling the depth of heavy water in the reacting volume. The moderator in the reacting volume is prevented from flowing to a well at the bottom of the reactor shell by a difference in pressure of helium gas. When it is necessary to stop the reaction quickly, the release of this pressure difference will permit the moderator to flow out rapidly. Under normal operation heavy water is pumped at a controllable rate from the well underneath, back into the reacting volume and there is a flow of heavy water from the reactor space through a weir near the bottom into the well underneath. The level of heavy water in the reactor is controlled by varying the rate at which it is pumped back into the core region; by this method the reactivity can be controlled over a range of about 0·3%. A wider range of control will be necessary to compensate for variations in the xenon poisoning when starting up after short shut down. This will be accomplished by controlling the temperature of the moderator, and will give a range of about 3% in reactivity. The temperature will be adjusted by regulating the flow of coolant to the heat exchanger through which the moderator returns from the well to the reactor core. These provisions for control are tied in with the ionization chambers that set the power level in a servo-circuit; and the whole system is automatic. It is designed to keep the saturation steam pressure delivered to the throttle of the turbine at the design value and keep the operating power of the reactor at the level of demand. In practice the reactor will be operated at a constant power; surplus steam will be dumped directly to the condensers as the electric loads vary.

Although the moderator and the coolant are at different temperatures their pressures will be equalized. The pressure will be only a few pounds per square inch above the saturation value. Surface boiling will occur on the cladding of the fuel rods where the maximum heat transfer rate is about 120000 B.t.u. ft $^{-2}h^{-1}$, but the coolant will be entirely in the liquid phase as it leaves the reactor. This is probably the first reactor to be constructed for operation without a high excess pressure. By keeping the pressure low the weight of

the pressure shell is reduced and the difficulties in its manufacture are simplified with considerable saving in cost. The reactor is not expected to produce electricity at competitive prices. It is intended rather to give Canadian industry experience in the design, construction and operation of a nuclear electric power station that would be helpful in reaching the goal of economic nuclear electric power soon after 1965.

Dr. J. V. Dunworth: Dr. Dietrich has spoken to you this morning on the physics of the fast reactors. Firstly I should like to take up myself one or two points connected with breeding.

It is often stated that a fast reactor is of great importance since it produces far more fuel than it consumes and it has been indicated in this session for example that in a practical design roughly one and a half atoms of plutonium can be made for each atom of plutonium which is consumed. It is well to realize, however, that present designs of fast reactor can burn fissile fuel at only a comparatively slow rate and it may well take five years or more to achieve doubling of one's stock of fissile material. At present prices for fissile material such a rate of gain is not of great economic significance, and it will be important, therefore, for the time being to consider the fast reactor on its merits as a power producing reactor rather than because a net gain of fuel is theoretically possible. I should like to point out also that one of the key problems in the development of the fast reactor system is associated with the processing of fuel. It is unfortunately not possible to dilute the fuel heavily as otherwise the neutron energy spectrum in the system becomes softer and the breeding gain deteriorates sharply. It may not be possible with the first designs of such reactors to burn more than 10% of the fissile material before reprocessing is required for metallurgical reasons. If, during this processing 5% of the fuel is lost then the whole of the breeding gain achieved by the fast reactor will be lost in the processing plant. I should not like my remarks on the fast reactor to be taken in any way as a criticism of this system but rather that one should be aware of factors which are well known in the atomic energy world but which tend to be grossly distorted by the popular press. One of the long term interests of the fast reactor, of course, is that it does afford a means of consuming all the ^{238}U in the world and it is not yet possible to see how this can be achieved in any other way.

It is theoretically possible to produce more fuel than is consumed by operating thermal reactors on the Th–^{233}U fuel cycle. The possible gain factors are small in that about the maximum which is likely to be achieved is the production of about eleven new atoms of fuel for ten which are consumed. It is possible to dilute the fuel in a thermal reactor and despite this small gain it may be possible to achieve doubling of one's stock of fuel in perhaps seven to ten years which is not much longer than that which is possible with a fast reactor system operating on the Pu–^{238}U fuel cycle. Once again, however, there is no major economic significance in this production of fissile material. Further, it will be quite economic to operate the Th–^{233}U system even if a slight loss of fuel occurs, since the cost of feeding in small quantities of ^{235}U or plutonium are quite acceptable and would not increase significantly the cost of electricity produced from a reactor system operating under such conditions. This point has been mentioned by the President in his opening address.

I am often told that it is wasteful to operate a Calder Hall system on natural uranium fuel and use only about $\frac{1}{2}$% of all the uranium atoms present in the natural uranium fuel. It is perfectly possible, however, to store the ^{238}U which remains against a future time when it can all be consumed in fast reactors and one need not, therefore, worry that one is sacrificing energy resources of future generations by using such a small quantity of fuel in present operations.

I have felt for some time that it is very important to take full advantage, in designing a reactor, of any safety mechanisms which can be built into the system by virtue of its nuclear properties and its transient characteristics. It is very difficult to make calculations from first principles on this subject and one must, therefore, envisage a practical experimental program which may ultimately provide the basis for a theoretical study. For this reason I was very anxious that Dr. Dietrich should present at this conference films showing the preliminary experiments which have been undertaken in the United States and we must all be very grateful to him for acceding to our request.* This whole problem, of course, is related particularly to the more advanced types of reactor.

I may end my remarks perhaps by mentioning that nuclear reactors will tend to have high capital costs and low fuel costs and are, therefore, ideally suited to operate on 100% loading. This makes it even more important to try to find some cheap method of obtaining energy storage in such a way as to meet the peak electricity load. It may come as a surprise to some that a good location for a nuclear power station is probably at Niagara Falls where it would be used to pump the water back up the falls, so that it could be used over again to provide the peak electricity load in the Province of Ontario.

Mr. P. W. Mummery: Assessments have been made at Harwell of the properties of thermal reactors operating on the uranium plutonium fuel cycle. It has been assumed that the cycle is operated with a feed of natural uranium, $E_f = 1$. Some of the uranium removed from the reactor is rejected. After extraction of fission products the remainder of the uranium together with the plutonium is mixed with the feed of natural uranium to provide the new charge for the reactor.

Under the conditions existing at equilibrium, that is when the isotopic ratios have reached steady values, it is possible to express the uranium utilization achievable in such a cycle as a function of the properties of the reactor when it is designed to operate initially with enriched uranium. In particular, the influence of conversion factor, C_0, enrichment, E_0, net fast neutron yield $\varepsilon f/(1 + K^2L_s^2)$, and neutron temperature can be evaluated.

The effect of the different variations with energy of the cross-sections of the various isotopes has been taken into account assuming that the neutron spectrum has the form of a Maxwellian changing to a Fermi $1/E$ spectrum at an energy of a few kT.

Fig. 4 on p. S 4 of this Supplement shows the effect of moderator temperature on the uranium utilization achievable.

Mr. G. R. H. Geoghegan: The distribution of fission products from a large number of fissions and the average number of neutrons per fission are normally regarded as immutable. However, all fissions are not alike and there would be a great advantage if by any means the distribution could be biased in a favourable way—for example, the reactor physicists would prefer a preponderance of fissions giving a large number of neutrons and the chemists would like to avoid the production of certain undesirable fission product elements. Is there any possibility of altering the distribution in practice?

* See p. S 107 of this Supplement.

Prof. H. W. B. Skinner: I do not believe that such a thing is possible.

Mr. C. N. Watson-Munro: Referring to Dr. Laurence's remarks on the Canadian approach, I should like to mention that Australia finds herself in a somewhat similar position, i.e. we have no urgent need for nuclear power but have a long term interest beginning in 10 to 15 years. If we assume that either (*a*) one of the thermal breeder types of systems operating on the thorium cycle or (*b*) a fast reactor is adopted, we need to be able to supply the capital fissile material. This will be provided by a diffusion plant, or by a natural uranium system such as the British gas cooled reactors or the Canadian heavy water one.

In Australia we are tending to think in terms of meeting this need of capital investment of fuel for the so-called Stage 2 or Stage 3 systems, by the use of PIPPA type reactors with part of the PIPPA capacity, if necessary, used as plutonium–^{233}U convertors.

Dr. J. R. Dietrich: I should like to ask Dr. Laurence whether my understanding, that a major part of the reactivity control in the reactor is accomplished by adjustment of moderator level, is correct. If so, I should like to ask him whether he anticipates any unusual difficulty in measuring and controlling this level under pressurized conditions.

Dr. G. C. Laurence: Direct control of reactivity is by adjustment of moderator level. When this results in a low moderator level, as will happen when starting after a long shut down, the flow of coolant to the moderator heat exchanger will be reduced automatically and the temperature of the moderator will rise slowly tending to reduce the reactivity. The moderator level will then rise to compensate until the normal range of operating levels, within a few inches of the top of the vessel, is reached.

We have tried to foresee any possible difficulties of operating under pressure. It was thought that helium dissolved in the heavy water might give trouble when the pressure was changed, but experiments have shown that this is unlikely.

SESSION 3. LECTURES

Chairman: J. V. DUNWORTH, C.B.E., Ph.D., F.Inst.P.

Experimental reactor physics

By P. W. MUMMERY, M.A., Atomic Energy Research Establishment, Harwell, Berkshire

The successful design, construction and operation of nuclear reactors requires detailed information concerning the interaction of various nuclear radiations with matter. In principle this can be obtained from a knowledge of fundamental atomic and nuclear properties combined with mathematical analysis. In practice a considerable number of integral measurements are required. These experiments measure combinations of various fundamental atomic and nuclear properties and the predictions of the mathematical analysis.

The detailed information required is outlined and various integral measurements are discussed together with their correlation with fundamental data and with various mathematical models. Consideration is given to the use of sub-critical, and low-power and high-power critical reactor assemblies in this field.

INTRODUCTION

The first nuclear reactors operated with naturally occurring uranium as the fissile material. Consequently the primary aim of the early reactor physics research was to ensure that such a reactor could be made critical and could be operated as a heat generating device. This was possible only with lumped fuel elements, with a reactor in which the neutron spectrum consisted mainly of neutrons in thermal equilibrium with the reactor materials and with a suitable moderator such as heavy water or pure graphite.[1]

Development of the nuclear reactor industry now requires that reactors should be designed for a variety of specific purposes. Examples are the production of electricity, of man-made fissile materials, of radioactive isotopes or of a high intensity of neutrons. In addition many more degrees of freedom have been introduced into the choice of reactor designs. The availability of man-made fissile materials, ^{235}U, ^{233}U, ^{239}Pu and ^{241}Pu enables the reactor designer to obtain many different critical systems. The effects of different coolant and structural materials, different neutron spectra in the reactors, dispersion of the fuel elements and the use of thorium as a fertile material must be evaluated.

The optimum design of reactors for specific purposes depends on many variables. It is the function of a reactor physics research programme to provide the physics data on which such a design can be based.

BASIC PROBLEMS

There are three basic problems which have to be solved in providing the reactor designer with the information he requires. The conditions for criticality for the system must be evaluated as a function of the composition and size of the system, of the concentration and disposition of the materials in the system, of temperature and of the power at which the system operates; all taking into account long-term effects due to the changes in the type of materials present resulting from neutron capture. The dynamic characteristics of the system and the rate of heat production at all points in the system must also be known as a function of all the variables.

The criticality condition

The critical condition can be expressed by the continuity equation

neutron yield = neutron absorption + neutron leakage

The solution of this equation requires that basic nuclear data are available; neutron cross-sections and neutron yields as a function of energy for processes which are important. In principle, a complete knowledge of all the relevant data would enable the neutron distribution in space and energy to be calculated together with the total neutron yield, absorption and leakage for any particular system. Such a calculation would be numerical and would require an electronic computing machine. In practice such complete data are not yet available nor is the accuracy of measurements of fundamental nuclear data sufficiently accurate for this purpose. For example, an uncertainty of 2% in the neutron yield per neutron absorbed in ^{235}U will lead to an uncertainty of a factor two in the critical size of a typical natural uranium, graphite moderated reactor.

The method adopted, therefore, is to make use of a selection of fundamental nuclear data which is available, combine this with some simple mathematical model of the behaviour of neutrons in the system and correlate the predictions from this procedure with integral experiments. Integral experiments measure combinations of nuclear data and the predictions of mathematical models. One important integral experiment is the determination of the state in which the criticality condition is satisfied.

It is usual to treat some of the nuclear data as unknown in this correlation and to determine values which fit the experimental results. A semi-empirical method of calculation can then be obtained which will agree with experiments and reasonable confidence can be attached to this if the nuclear data determined agrees with other integral measurements or with the fundamental measurements.

Reactor dynamics

Reactor dynamics relate to the changes in neutron intensity and distribution which occur when the criticality condition

is not satisfied. Again, in principle the problem can be solved so long as adequate fundamental data and computing facilities are available but in practice integral experiments are necessary. Such an integral experiment is the observation of the rate of increase and decrease of neutron intensity in a low-power critical system following the removal and addition of material which absorbs neutrons. A more elaborate experiment of the same type could include the effects of power and temperature transients with a variety of coolant conditions as in the case of the BORAX experiments[2] conducted at the U.S.A.E.C. Reactor Testing Site at Arco in Idaho.

Rate of heat generation

The distribution of the rate of heat production in the reactor is required so that an adequate heat dissipation circuit can be installed. Heat is produced as a result of the exchange of energy between the various energetic radiations and particles, present in the reactor, and the reactor materials. Therefore the distribution, origin and fates of such radiations and particles must be known. This problem is particularly important if the temperature of reactor materials must be limited and an example of this is the heat production in the reflector and shield. For a power generating reactor the distribution of the rate of heat generation in the core is vital since this will affect the overall coolant temperature and consequently affect the heat exchanger and turbine performance.

Again, integral measurements in appropriate assemblies of materials are required. For the core, the main source of heat is due to fission fragments from the fuel elements and so a distribution of fission will be most valuable. In the reflector and shield, however, appreciable contributions arise from γ-radiations and as the sources of γ-rays are primarily due to neutron capture the problem is more complicated.

INTEGRAL EXPERIMENTS

Reaction rates

It is possible to measure directly in a reactor assembly the relative reaction rates for any material which yields a detectable product of the process. If it is known that this process accounts for the whole of the neutron absorption cross-section and if such a measurement is possible with reactor materials, then the relative neutron absorption rates can be obtained for insertion into the equation for the criticality condition. In practice the neutron absorption in materials with a $1/v$ absorption cross-section can be measured using foils or boron counters, neutron fissions can be measured with fission chambers or by detecting the fission products from foils, neutron absorption in ^{238}U can be measured by detecting the ^{239}U 23 minutes half-life β-decay or the 100 keV X-ray from ^{239}Np. In general, foil techniques are more difficult to apply but they introduce fewer perturbing effects into the reactor assembly.

Examples of this kind of integral experiment are shown in Figs. 1 and 2. Fig. 1 compares the observed reaction rates in ^{239}Pu and ^{235}U relative to that of ^{10}B, and the expected rates calculated from cross-section data assuming that the neutron spectra were Maxwellians. The measurements were made in a thermal neutron distribution in a block of graphite which could be heated to 400° C.[3] Fig. 2 shows the relative reaction rates for a $1/v$ absorber as a function of position close to the central fuel element in a reactor assembly.[4] This is the so-called fine structure of the flux distribution which is due to the heterogeneity of the system.

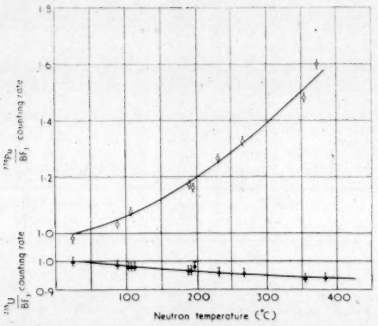

Fig. 1. Comparison of the observed reaction rates in ^{239}Pu and ^{235}U relative to that of ^{10}B. The ^{239}Pu and ^{235}U chambers were at graphite temperature

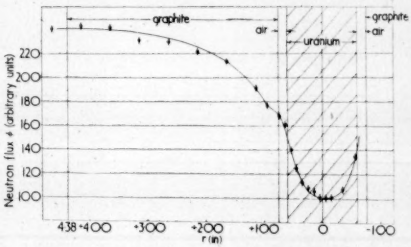

Fig. 2. Typical fine structure flux distribution

For neutron fluxes which are sufficiently high, as in a power reactor, it is possible to measure reaction rates by chemical or mass spectrometer analysis of irradiated materials. For example, chemical analysis of irradiated natural uranium will determine the relative plutonium content and its distribution through the sample. Mass spectrometer analysis will determine the relative amount of the various uranium and plutonium isotopes. From these results it is possible to reconstruct the reaction rates taking place during the irradiation.[5]

Neutron spectrum

The reaction rates are integrals of cross-section and neutron flux taken over space and energy. More detailed correlation with theory would be possible if the neutron spectrum were known. Indirect measurements can be made by using a variety of detectors and observing reaction rates. More direct measurements are desirable in at least a few cases as a check.

The neutron spectrum can certainly be observed by applying standard velocity selector techniques to neutron beams

extracted from reactor assemblies.[6] An alternative technique is to make use of a pulsed neutron source instead of the reactor neutron source thus avoiding the high average neutron intensities associated with the latter. Unfortunately, this technique is suitable only for the near thermal and thermal parts of the spectrum and then only for an assembly in which the slowing down time of fission neutrons is small.

Typical results[7] showing the difference between the neutron spectrum inside a fuel element and that in the moderator are shown in Fig. 3.

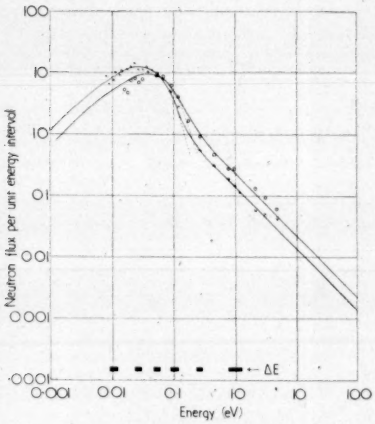

Fig. 3. Typical neutron spectra

○ spectrum in fuel.
× spectrum in moderator.

Neutron leakage

Neutron leakage can be defined in many ways. The important point is that a statement concerning neutron leakage also requires a statement concerning the system from which the neutrons are leaking. For example, leakage can be referred to the reacting core, the core plus blanket, the core plus reflector or the core plus reflector plus shield. The relative neutron losses as between absorption and leakage will therefore depend upon the definitions adopted.

In principle therefore a survey of reaction rates around whatever system has been defined will give the neutron leakage. In practice such a procedure is rarely carried out unless the system concerned is very small.

Criticality experiments

A critical experiment is essentially a reactor assembly in which some parameter can be varied until criticality is reached. The usual method is to increase the size of the reacting core. In terms of the continuity equation referred to previously, and defining the reacting core as the system considered, this procedure is equivalent qualitatively to decreasing the neutron leakage until the criticality condition is satisfied. The interpretation of this observed criticality condition in terms of mathematical models requires that detailed neutron distributions in space and energy should be observed.

It may also be possible to determine parameters, related to criticality, from sub-critical experiments. A typical mathematical model, the multi-group model, leads to the statement that the neutron distribution in a homogeneous reactor will be independent of energy at sufficient distances from boundaries and will satisfy the differential equation[8]

$$\nabla^2\phi + B^2\phi = 0$$

For a sufficiently large system therefore B^2 will determine the critical size; the boundary conditions applied are that for a bare system no neutrons enter the system from outside. In a sub-critical system a neutron distribution can be established by providing an external neutron source and the neutron distribution observed will then be related to B^2 and can be obtained by applying the appropriate boundary conditions. Such measurements in sub-critical reactor assemblies are called exponential experiments[9] and they have played a considerable role in reactor physics research programmes.

Reactor perturbation measurements

The effect upon reactivity of inserting or removing material from a low-power critical assembly is a sensitive integral experiment which requires only small samples of material, perhaps a few grammes. For a non-productive neutron absorber with a high-absorption cross-section the effect observed will be primarily a measure of the neutrons absorbed. In general, however, the effects of the modification of the neutron spectrum due to scattering and slowing down in the sample will also be important. In the case of fissile material the effect observed will be modified by the neutrons produced after capture.

Diffusion and slowing down

The characteristics of the behaviour of neutrons in various non-multiplying media can be examined by integral experiments which observe the neutron distribution as a function of energy and space arising from neutron sources of different energy. The mean square distance which neutrons travel during their conversion from one state to another, for example, fission neutrons to absorption as thermal neutrons, can be determined and used in the criticality condition. These distances are of primary importance in determining neutron leakage. In more general theory the form of the distribution observed can be used directly in the mathematical analysis.

Pulsed source experiments

Reference has already been made briefly to the use of a pulsed neutron source in determining reactor spectra but other measurements can be made. This technique differs from those mentioned previously in that instead of observing the spatial distribution of neutrons in a steady state, the time distribution of neutrons is observed in a non-steady state. Similar parameters can be observed and considerable success has been achieved with measurements of diffusion area, slowing down area, and thermal transport mean free path in non-reproducing media.[10] The method is being applied to sub-critical reactor assemblies.

TYPICAL CORRELATIONS

A specific example of the correlation between fundamental and integral measurements and mathematical models is that of establishing methods of calculation for low-power graphite-moderated natural-uranium lattices. Integral experiments

with such reactor assemblies are possible. Prediction of the changes which will occur after prolonged irradiation of natural uranium is also required. In this case use of reactor assemblies would require extensive facilities for handling highly radioactive material. A considerable amount of data can be obtained, however, from perturbation experiments.

Natural uranium graphite lattices

Mathematical model. The standard two group critical equation is used

$$\eta\varepsilon pf = (1 + B^2L^2)(1 + B^2L_s^2)$$

The neutron yield from ^{235}U and ^{238}U, $\eta\varepsilon$, is primarily dependent upon the fuel element shape and size. The fraction of fast neutrons which are captured in the ^{238}U resonance, $1 - p$, is capture additional to that incurred by a $1/v$ variation of the ^{238}U absorption cross-section. This factor p is primarily dependent upon the moderator to ^{238}U atomic ratio and the effective uranium resonance integral, the latter being dependent upon fuel element shape and size. The fraction of neutrons captured in ^{235}U, assuming ^{238}U to be $1/v$ absorber is f. L^2 and L_s^2 are the two group characteristic areas, diffusion area and slowing down area respectively, these being primarily dependent upon the diffusion area and the relative neutron capture in the moderator, and the slowing down area in the moderator respectively. B^2 is the geometrical factor for criticality, the parameter in the differential equation

$$\nabla^2\phi + B^2\phi = 0$$

Experimental procedure. Series of exponential and critical experiments enable B^2 to be determined and slowing down area measurements allow L_s^2 to be calculated. Reaction rate measurements allow f to be determined and in conjunction with diffusion length measurements in the moderator the value of L^2 can be found. With the model adopted and the materials concerned it is convenient to consider only the reaction rate for $1/v$ or near $1/v$ absorbers. Essentially then the measurement of these reaction rates can be interpreted in terms of fundamental cross-sections and the fine structure variation of neutron flux between the uranium lumps and the moderator.

From these measurements $\eta\varepsilon p$ is determined. By carrying out series of experiments in which for a given fuel element assembly the lattice spacing is altered, it is possible to determine $\eta\varepsilon$ and the effective uranium resonance integral for that fuel element. Repetition of these measurements with different shapes of fuel elements will give η, and the dependence of ε and the effective uranium resonance integral upon fuel element shape and size.

Results of correlation. It is assumed that

$$p = \exp\left[-\frac{Nu(\int\sigma_u dE/E)_{eff}Vu}{N_m\sigma_{sm}\xi_m V_m}\right]$$

Then

$$\log_e\frac{(1 + B^2L^2)(1 + B^2L_s^2)}{(1 + \delta\varepsilon)f(1 + \delta\eta)} = \log_e\eta_0\varepsilon_0 - \frac{Nu(\int\sigma_u dE/E)_{eff}Vu}{N_m\sigma_{sm}\xi_m V_m}$$

where $\delta\eta$ and $\delta\varepsilon$ are calculated corrections which allow for the influence of neutron spectrum and lattice pitch upon η_0 and ε_0 which are characteristic of a pure Maxwellian neutron spectrum and a single fuel element respectively.

Fig. 4 shows the fit which is obtained for three different fuel elements, and Fig. 5 shows that the values for the uranium resonance integral can be expressed in the form

$$(\int\sigma_u dE/E)_{eff} = A + B(S/M)$$

where A and B are constants and S/M is the ratio of surface to mass in the fuel element. The values for $\eta_0\varepsilon_0$ are consistent

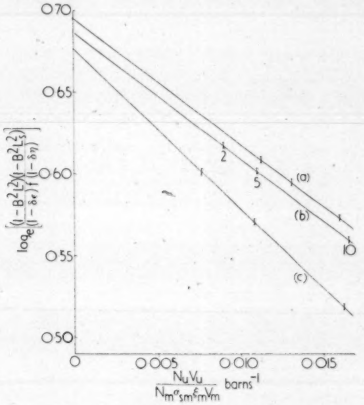

Fig. 4. Deduction of $\eta_0\varepsilon_0$ and $(\int\sigma_u dE/E)_{eff}$

(*a*) rod 1·20 in. diameter.
(*b*) rod 1·00 in. diameter.
(*c*) tube 1·64 in. outer diameter, 1·30 in. inner diameter

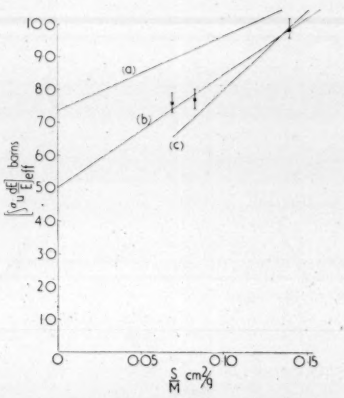

Fig. 5. Deduction of surface to mass formula

(*a*) Creutz's formula.
(*b*) $(\int\sigma_u dE/E)_{eff} = 5{\cdot}0 + 35\,(S/M)$.
(*c*) Crocker's formula.

with conventional calculation of the dependence of ε_0 upon fuel element shape and size.

With the values deduced from such a correlation which is described in more detail in Geneva Paper P429 it is possible to predict the results of a considerable number of lattice designs to an accuracy of $\pm\frac{1}{4}\%$ in reactivity.

The values reported at Geneva have been revised. Values of ε_0 and $(\int\sigma_d dE/E)_{eff}$ still agree reasonably well with reaction rate measurements although it is difficult to achieve high accuracy with the latter measurements. The value of η_0, 2·03, is now in good agreement with other measurements of this quantity, so that reasonable confidence can be attached to this correlation. It is of interest to note that the increase in η_0, of 0·09, is due primarily to changes in the basic nuclear cross-section data.

Long-term reactivity changes

The irradiation of natural uranium in a power reactor results in the destruction of ^{235}U yielding primarily fission products and ^{236}U, and the destruction of ^{238}U yielding primarily ^{239}Pu and fission products. The neutron cross-sections are such that appreciable quantities of other plutonium isotopes ^{240}Pu and ^{241}Pu are quickly built up in the reactor. The effect of these changes occurring as the reactor operates is that initially reactivity will fall due to the formation, as a fission product, of ^{149}Sm which has a high neutron absorption cross-section. The reactivity will then rise due to the replacement of ^{235}U by ^{239}Pu, if the amount of ^{239}Pu produced is sufficiently great, but will subsequently fall due to the formation of ^{240}Pu and the continuing production of fission products. The formation of ^{241}Pu which is fissile will decrease the rate of this fall. Such a curve is shown in Fig. 1 on p. S 2 of this Supplement.

The reactivity of a uniformly irradiated reactor will then follow this course. However, by suitable adjustment of the distribution of irradiation levels of the various fuel elements a steady state can be established in which the reactor remains critical with fuel being continuously fed into and rejected from the reactor. The irradiation level which can be achieved with each fuel element will then be approximately that which gives equal areas above and below the zero reactivity axis.

As in the case of the initial criticality condition this curve cannot be predicted sufficiently accurately from fundamental nuclear data. Furthermore, an accurate knowledge is required of the initial neutron fates and origins. With a knowledge of initial critical size and fundamental nuclear data only, the irradiation level which can be achieved with a typical natural uranium reactor is uncertain by $\pm 2\,000$MWD/t (approximately).

A number of integral experiments enables this uncertainty to be reduced. Measurements of the effectiveness of the various materials in reactor spectra by the use of foil and fission chamber techniques, and the chemical and mass analysis of natural uranium irradiated in a power reactor are all relevant. The most important measurement, however, is that of the net change in reactivity with irradiation due to the changes in composition. Natural uranium is irradiated in a variety of forms and conditions in a power reactor and then the changes in reactivity observed in perturbation experiments in a low-power-critical assembly in which a variety of neutron spectra can be provided. In this way semi-empirical methods can be established which will reduce the uncertainty in the estimates of long-term reactivity changes.

USE OF REACTOR ASSEMBLIES

It is convenient to consider the various reactor assemblies, which can be used, in four categories. These are sub-critical, near-critical, low-power critical and high-power critical. This distinction is particularly relevant when considering the cost and manpower required to design, construct and use these assemblies.

Sub-critical assemblies

These assemblies are so small that no re-orientation of the materials used or likely changes in the environment would result in them becoming critical. An increase of one dimension of the assembly to infinite length would not in general make the assembly critical and many assemblies are less than 20% of critical size. Under these conditions, and once it has been checked that the assembly is appreciably sub-critical, no instrumentation or safety devices are necessary.

Many measurements are possible in sub-critical assemblies if the neutron intensity which can be established by means of a neutron source is sufficiently high. A flux of 10^9 to 10^{10} n cm^{-2} s^{-1} is required to observe adequately the form of the neutron spectrum in the thermal and near thermal energy region, 10^7 to 10^8 for conversion factors and fast fission factors, 10^5 to 10^6 for fine structure effects and 10^4 to 10^5 for exponential type experiments. For the higher neutron fluxes the neutron source must be a power reactor.

Near-critical assemblies

These assemblies may be so near critical that re-orientation of materials used or likely changes in the environment could make the assembly critical. Careful loading procedures are necessary to achieve the desired condition and a certain amount of instrumentation including safety devices for automatically reducing reactivity must be provided.

This type of assembly is particularly useful if the characteristics of the assembly are such that it is difficult to extrapolate from sub-critical to critical conditions. As the assembly is near critical it is sensitive to small changes and the effect of perturbation can be observed. This technique has been used to measure the effect of small temperature changes and of control devices.

Critical low-power assemblies

These assemblies are designed to be operated in the critical and super-critical condition. In addition to safety devices, control devices must be provided together with additional instrumentation. Shielding is essential.

The principal advantages of a critical assembly are that dynamic effects can be studied, the effect of relatively large non-uniform changes to the assembly can be determined, accurate perturbation measurements are possible with very small samples and accurate neutron distribution measurements can be made without the use of an external neutron source.

Critical high-power assemblies

An assembly of this kind is necessarily very expensive because of the power generated. Such an assembly is required in order to change the composition of materials used in reactors. The effects of these changes can then be observed by placing the materials in other assemblies. Ideally the environment in which such irradiations can be carried out and the materials irradiated should be variable over a wide range.

CONCLUSIONS

In principle, reactor physics problems can be solved by the combination of numerical techniques, using electronic computers, and a complete knowledge of the fundamental nuclear and atomic properties of materials used in reactors. In practice this situation is not likely to be reached for many years even with simple systems, and even then the accuracy likely to be achieved is too low for many practical reactor problems. As a result, integral measurements are necessary in reactor assemblies and in some cases, particularly in perturbation measurements, these integral measurements will lead to improvements in our knowledge of fundamental data.

The enormous variety of possible reactor designs and the materials which they may contain requires that investigations of this kind will be necessary for many years. An important feature is the necessity for correlation of a sufficiently wide range of experimental results with methods of calculation so that the reactor designer can make use of this data.

ACKNOWLEDGEMENT

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The zero-energy fast breeder reactor, ZEPHYR

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ZEPHYR is a small low-power fast reactor with a plutonium core surrounded by an envelope of natural uranium. No cooling is required since the maximum operating power is about 2 W. This reactor first diverged in February, 1954, and has been used to make a detailed study of the neutron physics of fast reactors. Of the many aspects investigated, particular attention has been given to a determination of the breeding characteristics of the system and the measurement of the nuclear parameters required for a theoretical interpretation of fast reactors.

Interest at Harwell in unmoderated reactors dates back to the end of 1947. It was known that conditions in thermal reactors were not favourable for breeding fissile materials, except possibly in the ^{233}U–^{232}Th cycle. On the other hand it was expected that, in fast reactors, the ratio of the fission cross-section to the radiative capture cross-section in the three main nuclear fuels would be much higher than in thermal systems and that consequently it would be possible to achieve "breeding factors" significantly greater than unity in piles of suitable design. By "breeding factor" is meant the ratio of fissile atoms (e.g. ^{239}Pu) burned in the reactor over a given period to the number of fresh fissile atoms generated by absorption of neutrons in the breeder material (e.g. ^{238}U).

A small group under Dr. G. C. J. Dalton, early in 1948, began feasibility studies on possible designs of a small high power fast reactor consisting of a core of plutonium surrounded by a breeding blanket of natural uranium. It was envisaged that such a reactor would have a core about 6 in. in diameter and 6 in. long, cooled by a sodium–potassium alloy and producing, perhaps, 2 MW of heat. At the time when these studies were in progress, of course, there was no immediate prospect of obtaining the necessary quantity of pure or nearly pure fissile material to build a fast reactor. Subsequently, the Industrial Group of the Atomic Energy Authority undertook the development of fast systems and the idea of building such a small high power reactor was abandoned in favour of the much larger fast breeder reactor, which is now under construction at Dounreay.

At Harwell in 1951 the emphasis of the fast reactor work was shifted from engineering studies to considerations of the physics of these systems. The decision was taken to build a very low power uncooled fast reactor which would produce neutrons at a sufficient rate to enable us to study the nuclear physics and kinetics characteristics of plutonium/uranium assemblies. A small working party met for the first time in July 1951 to consider the design of a Zero Energy Fast Reactor to be known as ZEPHYR and construction was completed about two years later. The difficult task of fabricating the plutonium fuel elements for ZEPHYR was completed by the Metallurgy Division in January 1954, and the first assembly became critical on Friday, 5th February, of that year.

It should be emphasized that the design of ZEPHYR did not bear any resemblance to a possible useful power reactor. Considerations of fissile material allocation forced us to work with a minimum amount of plutonium, concentrated into a very compact core with the breeder uranium almost entirely disposed in a surrounding blanket. This is not a practical layout since a power reactor would be required to produce of the order of a 100 MW of heat and this could only be done in a system having a core of about a hundred times the volume of that of ZEPHYR. Nevertheless it was considered that the small system which we planned would yield useful data on the breeding characteristics, kinetics, neutron energy distributions and other important properties of fast plutonium/uranium systems and we proceeded on

that assumption. While the experimental programme on ZEPHYR was in progress, however, ZEUS, a second fast reactor of more realistic dimensions, was being designed and constructed. The core of this reactor has about fifty times the volume of the ZEPHYR core and contains more than 200 kg of ^{235}U diluted with three times that amount of ^{238}U. It is a roughly full sized, low powered, uncooled version of the Dounreay fast reactor and was constructed with a view to determining the nuclear characteristics of that system.

DESCRIPTION OF ZEPHYR

Short descriptions of ZEPHYR have already appeared in the literature[1,2,3] but some of the main features will be outlined in the present paper for the sake of completeness.

The reactor ZEPHYR consists of a core, which contains all the fissile material, surrounded by an envelope of natural uranium which is usually termed "the blanket." The fuel, plutonium, is in the form of rods canned in nickel, each fuel rod consisting of two halves 3 in. long by 0·22 in. in diameter. One end of each half has a copper cap over it and the two halves are joined together by means of a low melting point solder (~140° C) between the caps. The fuel elements are mounted vertically in the reactor and the upper half-rod in each is secured to the nickel can, the lower half-rod hanging freely. In the event of an accidental power excursion the solder would melt, allowing the lower half-rod to drop about a centimetre to the bottom of the can, thereby creating a gap of this length at the centre of the element. The creation of such a gap in an inner core element would cause a drop in reactivity of 0·025%. This fusible link is intended as an emergency shut down device to prevent melting or even evaporation of the plutonium fuel in the event of an uncontrolled increase of power.

The nickel-clad plutonium elements are mounted in vertical channels in a cylindrical core matrix (Figs. 1 and 2). There are 349 such channels in a regular hexagonal array each running parallel to the axis of the matrix. The matrix is approximately 15 cm in length and diameter. In the first core assembly it consisted of natural uranium so that the constitution of the loaded region of the core was as follows:

plutonium (density 15·8 g/cm^2)	48% by volume
uranium (density 18·7 g/cm^2)	33% by volume
nickel	10% by volume
copper	2% by volume
air	7% by volume

A second core assembly was made up of thin stainless steel tubes, air spaced, in a rather closer hexagonal array than that of the uranium matrix, giving a core constitution:

plutonium	52% by volume
nickel	11% by volume
copper	2% by volume
steel	6% by volume
air	29% by volume

The blanket of ZEPHYR consists of an inner hexagonal pillar about 1 m in height and 22 cm between opposite faces, with the core situated symmetrically in the central region (Fig. 2), and an outer tank of uranium rods packed so that about 80% of the volume is taken up by uranium. The whole assembly forms, roughly, a right cylinder of just under 1 m in length and diameter, with its axis vertical. The inner pillar is in two movable sections, the portion immediately above the core may be raised to give access to the core, while the lower section is fixed forming the bed for the core, the control rods and shut-off block. The latter is a cylindrical block of uranium 15 cm in diameter and 40 cm long which may be moved up or down through a channel through the lower pillar section immediately below the core. The control rods are six in number and consist of 2½ cm diameter uranium rods 45 cm long which move in vertical channels symmetrically

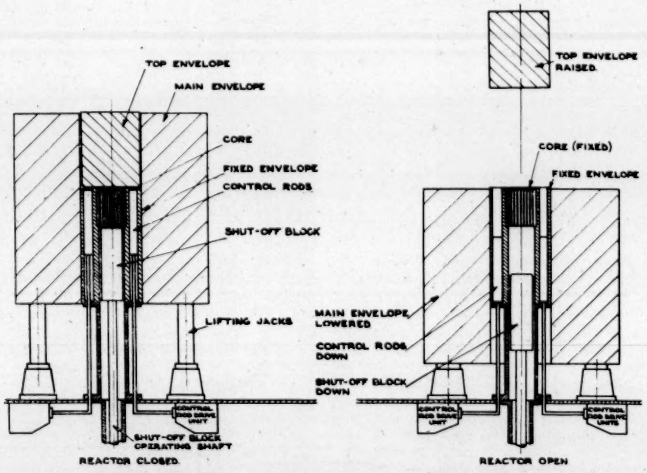

Fig. 1. Schematic diagram of ZEPHYR showing operating and loading position

arranged around the corners of the hexagon. When the rods are fully withdrawn they leave empty channels 15 cm long adjacent to the core.

The outer tank of uranium may be raised or lowered on hydraulic jacks. When it is in the raised position, the core is centrally situated with respect to the tank while in the lowered position the top of the tank is level with the top of the core, thereby ensuring that the latter is readily accessible.

The shut-off block controls a large amount of reactivity, namely 2 to 2½%, by variation of the neutron reflexion from the blanket into the core. When it is lowered, the large hole left below the core substantially increases the probability

nature of the core assembly). The control rods are moved on small motorized carriages to which they are attached by means of electromagnets. If the currents in the latter are interrupted as a result of accidental or deliberate trip signals, then the rods separate from the carriages and fall out freely under gravity. Only two of the six rods are used for fine control of the reactor, the remaining four act as additional shut down devices and must be fully inserted before the reactor can be made to diverge.

The measurement of power level in ZEPHYR is accomplished by means of fission chambers.[4] These employ ^{235}U as a fissile material and measure the fast neutron fluxes

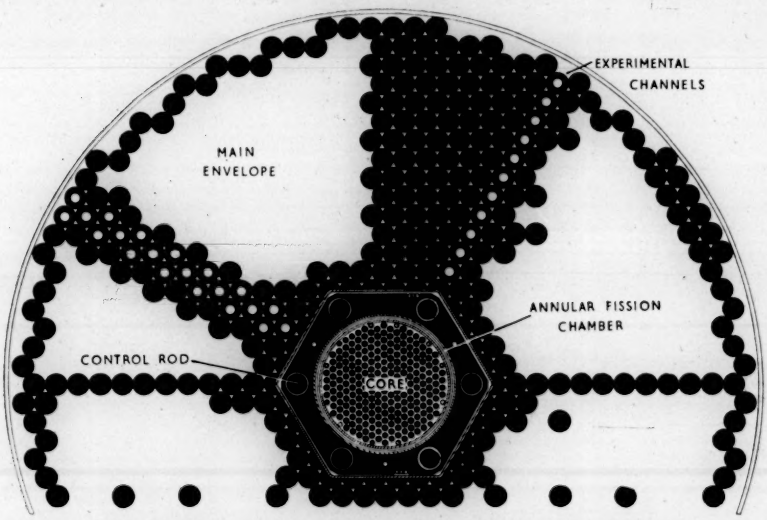

Fig. 2. Horizontal section through core and envelope of the reactor ZEPHYR

that neutrons may leak from the core. The loading of fuel in the core is always such that the reactor may not be made critical unless the shut-off block is fully inserted. Furthermore the outer blanket tank may not be lowered nor the upper pillar section raised to give access to the core unless the shut-off block is fully withdrawn. Conversely electrical interlocks ensure that the shut-off block may not be raised unless the upper pillar section is lowered and the outer tank in the raised position. The mechanism actuating the shut-off block is a hydraulic/pneumatic device, which will not be described here. Its specified performance is that it should provide slow insertion of the shut-off block, limiting the maximum rate of increase of reactivity to 0·01% per second and very rapid ejection, in the event of an emergency or manually initiated trip, removing about 2% of reactivity in about 200 ms.

The control rods also operate to vary the neutron reflecting power of the uranium blanket, but since they open or close relatively small gaps in the reflector they have a much smaller effect than the shut-off block, each control rod being worth about 0·125% in reactivity (depending, of course, upon the

in inner regions of the uranium blanket. Four channels are employed, one working with an annular chamber which completely surrounds the core of the reactor, the others connecting with chambers inside the main uranium tank near to the surface of the inner pillar and in the main plane of the core. The latter are held fixed with respect to the core when the tank is raised or lowered. All channels employ pulse counting with a very low resolving time permitting operation up to 10^5 pulses per second with accurately predicable losses. Two of the counting channels give a logarithmic presentation of their output and period meters are incorporated in these channels. Shut down trips are provided to operate from excess power level or excess rate of increase of power. These trips cause an interruption of current in the hold on magnets of the control rod carriages and also in a magnet which holds in a release valve of the shut-off block actuator.

ZEPHYR is normally operated at a power level of about 1 W. This figure refers to the power generated by fission and not to the total rate of release of heat in the core. Owing to the high rate of α-particle emission from the plutonium—a

normal core loading of about 15 kg produces some 30 W of heat, sufficient to maintain the core at a temperature of 20 to 50° C above ambient, depending upon the type of matrix which is employed. The shut-down power of ZEPHYR is governed by a RaBe neutron source, of strength 5×10^6 n/s, which is normally contained near the top of the main tank adjacent to the pillar, but is generally placed in the centre of the core during an assembly of the latter.

THE EXPERIMENTAL PROGRAMME ON ZEPHYR

The various methods of calculating the characteristics of fast reactors from basic nuclear data have been summarized in a recent paper by Codd, Shepherd and Tait.[3] Some of these methods are very powerful and could be relied upon to give excellent predictions provided that the available basic data is adequate. The need for building so-called zero energy reactors arises because of the difficulties involved in making accurate measurements of the fundamental nuclear parameters. One can cite, for example, the measurements of transport and inelastic scattering cross-sections which must be determined for a wide range of incident neutron energies and which involve difficult determinations of angular and energy distributions of emergent neutrons. Measurements on zero energy reactors and critical assemblies, covering a wide range of sizes and various constitutions of fissile, breeder and other materials, enable us to bypass many of the more difficult direct measurements of nuclear constants and to deduce best values for these to fit the observed behaviour of the experimental assemblies. The earliest work on the critical assemblies at Los Alamos and more recently on zero energy reactors and fast exponential experiments at the Argonne and at Harwell have gone a long way towards filling up the gaps in our knowledge of the physics of fast reactors.

The most important basic measurements which may be made in a zero energy system are as follows:

(i) critical size determinations;
(ii) measurement of neutron flux distributions;
(iii) determination of average nuclear cross-sections for various reactions, fissions, radiative capture, etc., in a wide variety of materials;
(iv) investigation of neutron energy distributions;
(v) observations on perturbations produced by small samples of various nuclear species;
(vi) observations on the effect of introducing neutron sources into various regions of a reactor;
(vii) measurements on the kinetic behaviour of systems and upon delayed neutron emission;
(viii) determination of the neutron economy and breeding.

All the aspects listed have been the subjects of experimental investigations in ZEPHYR. The results of some of these studies and their interpretation will be discussed in the last part of this report. It is worth while to precede these discussions, however, with an outline of the experimental techniques which are employed and the limitations which these impose upon the accuracy of the measurements.

THE MEASUREMENT OF NEUTRON FLUXES, REACTION RATES AND NEUTRON ENERGY SPECTRA

Fast reactors—even the largest—are comparatively small and an essential feature of any device which is introduced into such a reactor to make some particular measurement is that it too must be very small in order to cause a minimum disturbance to the behaviour of the system. Miniature ionization chambers and small foils and other samples were the main tools employed in the ZEPHYR studies and some attention had to be given to demonstrating that these could be employed in such a manner that they did not significantly modify the property which they were being used to measure.

Probably the most fundamental quantity that must be measured is the neutron flux and, since there are considerable variations between the neutron spectra in different regions of a fast reactor with a blanket of a strongly inelastic scatterer like uranium, it is particularly important that the detector used for this purpose should be relatively insensitive to neutron energy. Of all the neutron induced reactions which might be utilized in such a detector, the fission of ^{239}Pu is the most suitable. Over the full range of spectral variation found in ZEPHYR, i.e. between the centre of the air spaced core (specified earlier in this paper) and the outer regions of the uranium blanket, the fission cross-section for ^{239}Pu (averaged over either spectrum) varies by only 5%. A relatively crude knowledge of neutron energy spectrum is adequate, therefore, to determine the appropriate value of the ^{239}Pu cross-section to use in any particular circumstance.

Two alternative methods of using ^{239}Pu fission are open to us. The ^{239}Pu might be irradiated in the form of a foil and the subsequent fission product activity measured, or, alternatively, the material might be employed in a fission chamber. The first of these methods has the virtue that the foil might be made so small that it could not possibly disturb the neutron flux which it was employed to measure. On the other hand, the irradiation of samples and the subsequent determination of beta or gamma activities of fission products is a type of measurement which is particularly susceptible to experimental error. For this reason, and because it was found that small fission chambers could be used in the core of ZEPHYR without seriously modifying the flux, the latter have been used almost exclusively in ZEPHYR.

Among the nuclear reactions of particular interest in the ZEPHYR measurements, the fission of ^{235}U and ^{238}U and radiative capture in ^{238}U received special attention. Fission chambers were used in the first two of these and these chambers were intercalibrated with the ^{239}Pu chambers used in flux measurements. This intercalibration consisted of the determination of the effective quantity of the fissile material in each chamber by observing fission rates in known thermal fluxes and assuming values for thermal neutron cross-sections. The counting rates observed in these chambers when they are irradiated in a given fast neutron flux then indicates the ratios of the corresponding fission cross-sections averaged over the neutron energy spectrum in that flux. Uncertainties in the relative thermal fission cross-sections combined with the normal experimental errors set an upper limit of between 5 and 10% on the accuracy of cross-section measurements obtainable in this manner.

Radiative capture cross-sections in ^{238}U may be determined from observations of the ^{239}U or ^{239}Np activity induced in irradiated samples. In the ZEPHYR studies we have preferred to observe the ^{239}U activity despite its short half-life of 23 min, which makes it necessary to carry out the solvent extraction process (to separate fission product activities) in the space of a few minutes. Sources of error involved in this measurement lie mainly in the calibration of the ^{239}Pu fission chamber used for the flux measurement, in the calibration of the beta-counter used to measure the sample activity,—including the various corrections for self-scattering and self-absorption, and in the correction for fission product activities

which remain in the sample despite the solvent extraction. We consider that the best accuracy which we could claim in the determination of the ratio of $\sigma(n, \gamma)^{238}U$ to $\sigma(n, f)^{239}Pu$ is $\pm 7\%$.

The direct measurement of neutron energy spectra inside a diffusing medium is particularly difficult, especially in the energy region below 1 MeV. The analysis of proton recoils in photographic plates may be used to determine the spectrum above 1 MeV, but it is extremely laborious when the direction of the incident neutrons is unspecified. J. E. R. Holmes[1] has used proton recoil pulse analysis in fairly large proportional counters placed in the blanket of ZEPHYR and R. Batchelor has used a ^{3}He proportional-counter spectrometer to extend the range of measurement down to 100 keV. However, these spectrometers are so large that they could not possibly be accommodated in the core of ZEPHYR. The alternative of extracting a beam from given regions of the reactor and analysing the energy distribution of the neutrons in the beam has not been used so far, at Harwell, though this method has been used extensively by workers at the Argonne National Laboratory. There is a serious uncertainty in this method arising from the possibility of there being significant differences between the beam spectrum and that of the flux in the region from which the beam is extracted.

Indirect methods of deducing neutron energy distributions have been used for the purpose of analysing the ZEPHYR core spectra. For this, detectors based upon particularly energy sensitive reactions such as radiative capture [e.g. $^{197}Au(n, \gamma)^{198}Au$] which are very sensitive to lower energy neutrons and threshold reactions [e.g. $^{237}Np(n, f)$ and $^{32}S(n, p)^{32}P$] which are only sensitive to neutrons above certain minimum energies have been employed. The procedure adopted is to carry out measurements in any given region of the reactor with a large number of detectors whose energy response is known from reaction cross-section measurements with monokinetic neutrons and then to "invent" a spectrum which will give the best possible fit to the cross-section observed in that region.

REACTIVITY MEASUREMENTS, PERTURBATIONS AND DELAYED NEUTRONS

A wide field of measurements on ZEPHYR requires a knowledge of the reactivity scale of the reactor, i.e. it is necessary to know what reactivity changes are produced by given movements of control rods or by dropping the shut-off block, for example. The basic calibration is the determination of the reactivity change corresponding to, say, a centimetre movement of control rod. Other changes such as the insertion or ejection of the shut-off block can be related to this by observations of the relative changes in multiplication of the system which they produce when the reactor is sub-critical. Actually, the calibration of a ZEPHYR control rod is not uniform over the full extent of its traverse, a centimetre movement near the centre region being more effective than the same movement near either end of the stroke. So far as possible one attempts to utilize the central region rather than the extreme ends of the control rod movement when making sensitive measurements of perturbation.

The absolute reactivity calibration of ZEPHYR has been achieved by introducing a small sample of plutonium into a space at the centre of the core of the reactor and determining the control rod movement required to compensate the reactivity increment produced by the plutonium. Provided that the appropriate nuclear constants of ^{239}Pu and flux and fission rates throughout the reactor are known, the reactivity change produced by the sample can be calculated. For this purpose a one-group perturbation theory may be used.[5] According to this the change of reactivity $\delta\rho$ resulting from the introduction of a small sample of volume δV into a vacant space at some point r in the system is given by

$$\delta\rho = A[\beta - g(r)\alpha]\phi(r)^2\delta V \tag{1}$$

where $\phi(r)$ is the neutron flux at r, α and β are the one-group parameters of the sample material which in the case of a single nuclear species are defined as follows:

$$\alpha = N(\sigma_f + \sigma_c + \sigma_{str}) \tag{2}$$

$$\beta = N(\sigma_{str} + \nu\sigma_f) \tag{3}$$

N being the number of nuclei per unit volume, σ_f the fission cross-section, σ_c the effective radiative capture cross-section, σ_{str} the combined elastic and inelastic scattering component of the transport cross-section and ν the number of neutrons per fission in the sample.

The parameter $g(r)$ is the so-called perturbation function which in diffusion theory is given by

$$1 - g(r) = 3[j(r)/\phi(r)]^2 \tag{4}$$

$j(r)$ being the neutron current at r.

Finally, the quantity A is given by

$$\frac{1}{A} = \iiint p(r)\phi(r)dV \tag{5}$$

where $p(r)$ is the rate of production of neutrons due to fissions at r and the integral is taken over all regions in which fissions occur in the reactor.

The breakdown in simple one-group theory will be discussed later. It affects the calculations in so far as the degradation of neutron energy in an inelastic scattering collision changes its effectiveness or importance. One can interpret the effect as being equivalent to an absorption or emission of neutrons by adjusting the value of σ_c. This effect is relatively unimportant in the case of a fissile material owing to the predominance of the quantity $\nu\sigma_f$. The assessment of the quantity A, which is done by integrating fission rates (deduced from fission chamber measurements) over the whole reactor, is subject to some uncertainty with respect to the effectiveness of neutrons in the blanket of the reactor. However, for ZEPHYR, the core contributes about 90% to the value of A and a large measure of uncertainty in the blanket contribution is tolerable.

To eliminate any uncertainty in the value of the quantity $g(r)$, the sample of plutonium is introduced at the core centre where the neutron current is zero and in these circumstances $g(r)$ is unity and the expression for the reactivity change reduces to

$$\delta\rho = A(\beta - \alpha)\phi(0)^2\delta V \tag{6}$$

$\phi(0)$ is usually normalized to unity and the value of A is estimated on the basis of unit central flux.

The accuracy in estimating absolute reactivity scales by this method is between 5 and 10%. The values of the nuclear parameters used in the case of ZEPHYR are

$$\nu = 3{\cdot}1$$
$$\sigma_f = 1{\cdot}85 \text{ barns}$$
$$\sigma_c = 0{\cdot}20 \text{ barn}$$

giving

$$\beta - \alpha = 3{\cdot}7 \times 10^{-24} \text{ N cm}^{-1} \tag{7}$$

N being the density of nuclei of ^{239}Pu in the sample.

Having determined the absolute scale of reactivity it is possible to determine the effective delayed neutron fraction and relative yield of the different delayed neutron groups by two methods, viz:

(i) analysis of power level decay resulting from a known reduction in reactivity;

(ii) measurement of the steady reactor periods resulting from given increments or decrements of reactivity.

In the first case the reactor is run at a steady power level p_0 and then shut down by a rapid reduction of reactivity $\delta\rho$. If the time taken for this reduction is small compared with the shortest delayed neutron period, then there is a more or less instantaneous drop in power level to p_1 (the rate of drop is governed by the mean lifetime of a neutron which is $\sim 10^{-8}$ s in ZEPHYR), where p_1 is given by:

$$\frac{p_1}{p_0} = \frac{B}{B + \delta\rho} \tag{8}$$

B being the sum $\Sigma\beta_i$ of the fractions of neutrons in all the delayed neutron groups (i). Hence, if $\delta\rho$ is known B may be determined. Individual relative yields of neutrons may be obtained by analysing the subsequent decay curve of the power into a set of simple exponentials each of which corresponds to a given delayed neutron group. The actual relative yields and periods of the delayed neutrons differ from the observed yield and periods of the decay curve, because the neutrons from the delayed emitters that were in the system when it was shut down give rise to further delayed neutron emitters, so that, for example, fresh short period emitters are being generated long after the original ones have decayed to negligible levels. However, if the shut down reactivity $\delta\rho$ is several times as great as B, simple corrections may be applied to obtain the true periods and yields from the observed ones.

In ZEPHYR the measurement involved tripping the reactor so that the control rods fell out and the shut-off block was dropped. The total time involved in this process is about 0·2 s and as a result the lowest period delayed neutron group (period $\sim$200 ms) was masked and it was only possible to determine the yields of the five remaining group.

The second method mentioned above involves the use of the "inhour equation" and is so well known that it need not be discussed here. A third method involving the application of a known reactivity oscillation and the determination of the amplitudes of the resulting power level oscillations of the reactivity as a function of the frequency of oscillation has never been applied to ZEPHYR.

Extensive perturbation experiments on a wide variety of samples (elements and separated isotopes) were carried out in ZEPHYR by introducing the samples into the centre of the reactor and then into a region near the core boundary. In each case the resulting reactivity change was determined by control rod balance, i.e. by moving a control rod by a sufficient amount to restore the reactivity of the system to its original value. The sensitivity of the method is determined by two factors, namely the reproducibility of control rod positioning (the main source of irreproducibility being a slight lateral slop) and the sensitivity of the power level detectors, which limit the smallest drift in the reactor power that is detectable. In most of the work these two factors have limited the sensitivity to a minimum observable change of 5×10^{-7} in reactivity. Recently, using much more sensitive detectors it has been found possible to observe power drifts due to reactivity changes of 10^{-7} but the existing control rods do not permit us to exploit this sensitivity in the balance method. The limit of sensitivity of an oscillator method now in operation on ZEUS may be extended to about 5×10^{-8} in reactivity.

The reactivity change observed with a sample at the centre of the reactor core where $g(r)$ is unity, gives the value of $\beta - \alpha$ for that sample. In the case of fissile material samples this is given by

$$\beta - \alpha = N[(\nu - 1)\sigma_f - \sigma_c] \tag{9}$$

on the simple one-group theory, which reduces to $-N\sigma_c$ for non-fissile samples.

Measurement in a second position, where $g(r)$ is not zero, but is known, taken in conjunction with the value of $\beta - \alpha$ obtained from the central measurement enables one to determine the separate values of α and β, or in the case of non-fissile materials, the separate values of transport and effective capture cross-sections. The determination of $g(r)$ in a system may be made on the basis of the diffusion equation given above provided that one knows the neutron currents as a function of position. Neutron current is related to flux through the equation

$$j(r) = \left(\frac{\beta_r - \alpha_r}{\kappa_r^2}\right) \text{grad}\, \phi(r) \tag{10}$$

where α_r and β_r are the one-group parameters for the reactor medium at (r) and κ_r^2 is the Laplacian or buckling of the medium at (r).

It will be seen that some basic information is required before this method can be applied. If the value of $\beta - \alpha$ for ^{239}Pu is deduced from basic cross-section measurements then $\beta_r - \alpha_r$ can be obtained by a simple perturbation comparison of a ^{239}Pu sample and a small sample of the reactor medium. Once $\beta_r - \alpha_r$ is known, the Laplacian κ_r^2 can be deduced from the flux distribution, at least in principle. In practice, this is not a very feasible procedure unless the geometry of the system is very simple. The fact that the expression for $1 - g(r)$ contains $j(r)^2$ means that errors in estimating the neutron currents are doubled so that the accuracy of determining the perturbation function by this method is not very great. A better procedure to adopt is to take a standard sample of a non-fissile species for which σ_{str} is known as a function of energy and whose average value can be estimated, therefore, over the neutron spectrum in the reactor medium. The value of $\beta - \alpha$ for this sample can be obtained in a perturbation experiment by comparison with ^{239}Pu and hence α and β determined separately. A further perturbation measurement on the sample at the point r then enables us to determine $g(r)$.

The second procedure described in the previous paragraph was used to determine $g(r)$ values in ZEPHYR.

FLUX AND ENERGY DISTRIBUTIONS IN ZEPHYR

It is not proposed to give a detailed catalogue of the results of measurements in ZEPHYR but rather to discuss a few specific results which give a good picture of the physics of the system.

Fig. 3 illustrates the manner in which the counting rates in a number of different fission chambers and the rate of activation of a gold foil vary in going through the core and the solid uranium envelope above the core. The curves show the manner in which the neutron flux and energy spectrum varies through the core and envelope. If the cross-sections for the various reactions are normalized to unity at the core

centre then elsewhere they appear as in Fig. 4. (These curves all refer to the core assembly in the uranium matrix.) The variation in cross-section demonstrates that at the boundary of the core of the reactor the neutron spectrum is rather more degraded than in the centre, while, as one proceeds outwards, it becomes still more degraded, eventually tending towards an equilibrium in the outer regions of the blanket. The governing factor in this energy degradation is the process of

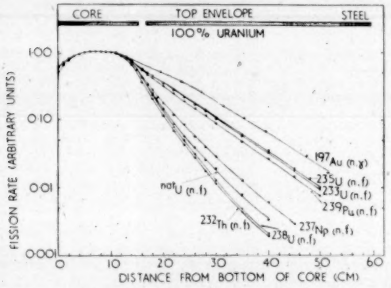

Fig. 3. Variation of fission rates through core and top envelope relative to centre of core

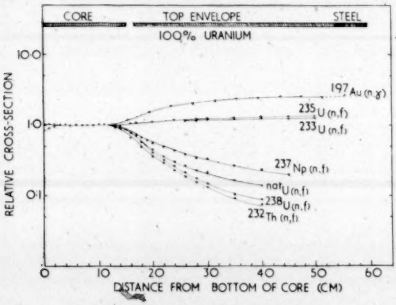

Fig. 4. Variation of cross-sections through core and top envelope relative to centre of core

inelastic scattering in ^{238}U, while the eventual equilibrium state which would be reached after passing through a large distance in the uranium blanket arises mainly from the replacement of high energy neutrons as a result of fissions in ^{235}U. The final degree of degradation is a function of the ^{235}U concentration, since only the ^{235}U fission is able to replace low energy neutrons by ones of high energy. Measurements in a thorium envelope revealed no tendency for the neutrons to reach a state of equilibrium, the spectrum far from the core becoming much more degraded than that in uranium, a result of the absence of any low energy neutron fission in thorium.

Neutron energy spectra for two cores (core I referring to the assembly with the uranium matrix and core II to that with the air spaced matrix) are shown in Fig. 5 in comparison with a fission spectrum. The core spectra were deduced by Holmes, Curry and Waldegrave[6] mainly from cross-section measurements in the manner previously discussed and, above 1·8 MeV, from the analysis of proton recoil tracks in tiny photographic plates exposed inside the core. It will be seen that above about 1½ MeV the forms of the core spectra are similar to one another and to the fission spectrum, but that there is a higher proportion of these high energy neutrons in the case of the pure fission distribution. Addition of more inelastic scattering material, viz. natural uranium, makes the core I spectrum more degraded than that of core II. It appears from these spectra that the average energy of neutrons in core I is 1·5 MeV and in core II 1·7 MeV, while that in the outer regions of the blanket is as low as 0·5 MeV. These

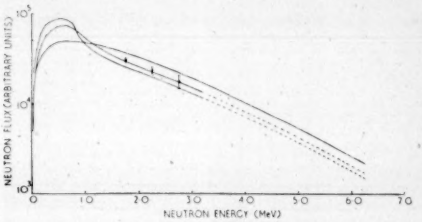

Fig. 5. Neutron energy spectra in ZEPHYR cores

data refer to the centre of the core. In the following tables a few cross-section values are listed for the two cores, for a fission spectrum in the centre of a ZEUS core assembly (25% ^{235}U + 75% ^{238}U) and in the ZEPHYR blanket as near as possible to the equilibrium region. The cross-sections chosen are $^{239}Pu(n, f)$, which is relatively insensitive to neutron energy above 10 keV, $^{238}U(n, f)$ which has a threshold in the region of 1 MeV and $^{197}Au(n, \gamma)$ which is typical of radiative capture processes and is most sensitive to low energy neutrons.

Table 1. *Reaction cross-sections in barns in various fast reactor spectra*

	Spectrum				
		ZEPHYR	*ZEPHYR*		*ZEPHYR*
reaction	*fission*	*core II*	*core I*	*ZEUS*	*blanket*
$^{239}Pu(n f)$	1·89	1·85	1·83	1·79	1·75
$^{238}U(n, f)$	0·28	0·21	0·18	0·091	0·01
$^{197}Au(n, \gamma)$	0·14	0·15	0·17	0·25	0·43

The extent to which neutron energies become degraded with increasing dilution of the fissile material by the breeder material is well illustrated by these figures, the threshold reaction falling very sharply and radiative capture increasing in a more gradual manner. The state of affairs in an actual fast power-breeder reactor would probably lie somewhere in the range between ZEUS and the ZEPHYR blanket provided that no great quantities of moderating material were present.

BREEDING

It has been pointed out in previous papers[1,3] that a high breeding ratio is achieved in ZEPHYR, the final values obtained after repeated measurements being

core I	2·0 ± 0·15
core II	1·9 ± 0·15

These high values may be attributed to the favourable ratio of fission to radiative capture cross-sections of ^{239}Pu in the high energy neutron spectra obtaining in these assemblies and also to the considerable bonus of neutrons resulting from fission in ^{238}U. The following balance sheet (Table 2) shows the distribution of neutrons between the various reactions which occur in ZEPHYR (core II) and compares the numbers with estimates of the numbers of neutrons produced based on the best available ν values for fast neutrons. It will be seen that the agreement between the columns is well within the quoted accuracy of measurement.

Table 2. *Distribution of neutrons between various reactions in ZEPHYR*[6]

reaction		*neutrons absorbed per 100 fissions*	*neutrons produced per 100 fissions*
$^{239}Pu(n, f)$	fissions	64 ± 5	198 ± 15
$^{235}U(n, f)$	fissions	10 ± 1	25 ± 3
$^{238}U(n, f)$	fissions	26 ± 2	66 ± 5
$^{239}Pu(n, \gamma)$	radiative captures	3 ± 1	
$^{235}U(n, \gamma)$	radiative captures	3 ± 1	
$^{238}U(n, \gamma)$	radiative captures	151 ± 10	
$Ni(n, \gamma)$	radiative captures	0·1	
Leakage		33 ± 10	
Total		290 ± 20	289 ± 20

The relative fission rates are deduced from fission chamber measurements; ^{238}U captures estimated from the activation measurements described above; the radiative capture in ^{239}Pu and ^{235}U are estimated from the reactor EBR-I cross-section data reported at Geneva[7]; leakage of neutrons from the outer blanket of ZEPHYR is deduced from flux distribution measurements.

In a large power reactor it is probable that a greater proportion of the breeder material will be introduced into the core to give a much larger volume and to reduce depletion effects. This will have the effect of degrading the neutron spectrum thereby increasing the radiative capture cross-sections. From the behaviour of ^{197}Au in Table 1 it would seem unlikely that the relative loss of neutrons by radiative capture in ^{239}Pu would increase by more than a factor of three. Provided that no other inelastic scatterer or light moderator were introduced into the system in significant quantities the fast fission rate in ^{238}U would be almost unaffected. It would appear from these facts that the breeding ratio in such a large system would not be seriously reduced from the high values observed in the case of ZEPHYR.

PERTURBATION STUDIES IN ZEPHYR

Perturbation measurements in ZEPHYR were carried out in both core assemblies, covering most of the natural elements, mainly by R. D. Smith and H. Rose. The results of these measurements were summarized at Geneva[1], the main features of these results being as follows:

(i) If one deduces capture cross-sections from measurements of perturbations produced by samples introduced at the core centre, on the basis of simple one-group theory, then the values so obtained are significantly greater than those obtained by activation measurements (where this is possible).

(ii) The effective capture cross-sections which are obtained from the perturbation measurements are much lower in the "magic number" regions than elsewhere and on the average the values for even-Z elements are less than those for odd-Z.

In order to pursue the last feature further Rose[8] has extended the measurements to a number of separated isotopes, and the results obtained so far indicate that those with odd numbers of nucleons A have substantially higher capture cross-sections than the even-nucleon elements. This effect could account largely for the difference between odd-Z elements (which are almost all odd-A) and even-Z elements in which most of the more abundant isotopes are of even-A.

The differences between the perturbation capture cross-sections and the true cross-sections signify that the one-group theory does not adequately explain the behaviour of fast reactors, such as ZEPHYR. This is not surprising in view of the dependence of most of the relevant nuclear cross-sections upon neutron energy. Because of this dependence the "importance" of a neutron in a reactor is a function of its energy, which can be changed as a result of elastic or inelastic collisions. Two effects enter into the behaviour of a perturbation sample at the centre of the ZEPHYR core. The first is the change of neutron importance as a result of energy loss in a scattering collision and the second is the fact that radiative capture selects neutrons which are of lower than average energy and whose importance therefore is not average.

It is worth noting that only a small variation of neutron importance with energy is sufficient to explain the ZEPHYR results. A qualitative interpretation requires that there is a minimum importance attached to neutrons in the middle regions of the neutron spectrum with a slight increase as one goes to higher energies (>1 MeV) and an increase also at lower energies. Inelastic scattering is a process which is transferring neutrons from the upper energy region to the middle region with a resulting loss of importance. Elastic scattering in hydrogen and deuterium on the other hand appears to transfer neutrons mainly from the medium region to the lower region with increase in importance. Results are shown for several species in Table 3. True absorption coefficients are shown as negative.

Table 3. *Central perturbation cross-sections*

species	*ZEPHYR core I perturbation activation (millibarns)*		*ZEPHYR core II perturbation activation (millibarns)*		*GODIVA*
^{1}H	+190	—	+256	—	+590
^{9}Be	+ 31	—	+ 46	—	+ 91
^{12}C	0	—	0	—	+ 33
*nat*B	−165	—	−116	—	− 85
^{23}Na	− 21	−1·0	—	—	+ 12
Fe	− 55	—	− 54	—	0
Br	−148	− 65	—	—	− 16
In	−305	−274	—	—	−156
^{127}I	−238	−160	—	—	− 66
^{181}Ta	−265	−232	—	—	− 98

This is not a complete list but it serves to illustrate the main features. In this list absorption effects are shown as negative while effects equivalent to neutron production are shown positive. As an interesting comparison some results obtained in the reactor GODIVA, the bare ^{235}U assembly at

Los Alamos, which were reported at Geneva[9] are also given. One can deduce from the GODIVA results that the net effect of both elastic and inelastic scattering is to increase the importance of neutrons. This indicates that the variation of neutron importance with energy in a ^{235}U system is appreciably different from that in a ^{239}Pu system. Qualitatively, at least, this can be explained by the differences between the energy variation of fission cross-section between the two isotopes (Fig. 6). In the case of ^{235}U the fission cross-section increases markedly with decrease of energy, whereas, in the case of ^{239}Pu there is an appreciable dip in this cross-section between 50 keV and 2 MeV.

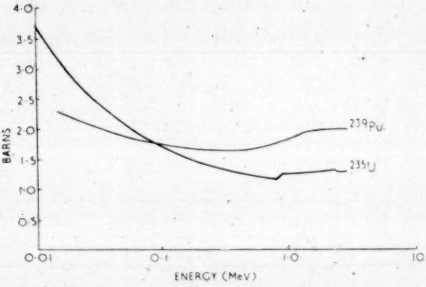

Fig. 6. Fission cross-sections in the MeV region for ^{239}Pu and ^{235}U

THE DELAYED NEUTRON FRACTION

Analysis of shut-down power decay in the manner discussed previously has yielded the following results for the delayed neutron fractions in the five longest groups.

On the other hand the effective yield of delayed neutrons from x is

$$\iiint E_d(r)B_x p_x(r)\phi(r)dV \qquad (12)$$

where $E_d(r)$ is the average value of the importance of delayed neutrons at r relative to average fission neutrons and B_x is the delayed neutron fraction of x.

In order to estimate the overall delayed neutron fraction, therefore, it is necessary to know the $E_d(r)$. Rose and Absalom[12] have made a survey with RaBe, mock fission and various photoneutron sources to determine the value of $E_d(r)$ as a function of neutron energy in the blanket of ZEPHYR. To do this they have placed various calibrated neutron sources in different positions in the blanket of the subcritical system and measured the output of neutrons from the reactor. If s is the source strength $E_s(r)$ the importance of the neutrons and $\delta\kappa = 1 - k$, k being the reproduction factor of the system, then the number of extra neutrons produced by the reactor when the source is placed at r is given by

$$N(r) = \frac{E_s(r)S\phi(r)}{\delta k}$$

The actual determination of $N(r)$ is not really necessary since in practice the sources are compared with a mock-fission source for which E is taken as unity. (It appears to be substantially independent of r.) A correction has to be made to allow for any change in δk resulting from perturbation by the source material.

The measurements have not been completed in so far as only RaBe and SbBe sources have been examined in the core. However, results suggest that the average value of E in the blanket of ZEPHYR should be about 0·7 for the energy spectrum of delayed neutrons. The value of E_d in the core has yet to be evaluated but it should not be less than 0·9 (a drop of neutron importance to this value in the middle energy regions is quite sufficient to explain inelastic scattering effects observed in the perturbation studies). If one takes

Table 4. *Relative yields and total delayed fraction* $\Sigma\beta_i$ *for 5 groups*[10,11]

group (i)	$t\frac{1}{2}$ (s)	*core I*	*core II*	^{239}Pu	^{238}U
1	55·6	0·025 ± 0·004	0·027 ± 0·001	0·031 ± 0·002	0·012 ± 0·001
2	22·0	0·229 ± 0·005	0·226 ± 0·001	0·304 ± 0·013	0·154 ± 0·005
3	4·5	0·341 ± 0·019	0·298 ± 0·015	0·317 ± 0·014	0·266 ± 0·017
4	1·5	0·405 ± 0·036	0·319 ± 0·016	0·293 ± 0·024	0·401 ± 0·032
5	0·43		0·13	0·055 ± 0·018	0·167 ± 0·033
		0·34 ± 0·01%	0·29 ± 0·01%	0·23 ± 0·02%	1·5 ± 0·17%

Fissions in ZEPHYR, as we have seen above, occur in the three isotopes ^{239}Pu, ^{238}U and ^{235}U and, since the delayed neutron fraction in ^{238}U is much higher than in ^{239}Pu, its contribution to the overall delayed neutron fraction is quite significant. In fact in a uniform infinite medium consisting of ^{239}Pu and ^{238}U the delayed neutron fraction would be about 0·50% with ^{238}U contributing more than half the neutrons. In ZEPHYR the situation is different because the distribution of fissions is not the same for the three fissile components and their neutrons are not of the same "weight" therefore. If we denote by suffix x a particular fissile component and take the importance of a fission neutron as unity, then the effective production rate from fissions in x throughout the system is

$$P_x = \iiint p_x(r)\phi(r)dV \qquad (11)$$

$E_d = 0·95$ in the core (which may be too high) and 0·7 in the envelope then the estimated delayed neutron fractions in ZEPHYR would be as follows:

B (5 groups) core I = 0·35%

B (5 groups) core II = 0·31%

which values are fairly close to those observed.

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DISCUSSION ON SESSION 3

Prof. O. R. Frisch: We have gone a long way since the days when we obtained some small bricks of ^{235}U in Los Alamos, to assemble into approximately spherical shapes and determine critical sizes: I am very much impressed with the power and flexibility of these new methods. Could you tell us something of your plans for the future? When you irradiate emulsions, what do you do to avoid overexposing them? Could you say a few words about the ^{3}He spectrometer which you mentioned?

AUTHOR'S REPLY

Dr. L. R. Shepherd: It is not possible to give a comprehensive answer to Professor Frisch's first question. However, in general, the work will involve studies of much larger systems than ZEPHYR. In these, the neutron spectra will be more degraded as a result mainly of inelastic scattering in materials which are used to dilute the fissile component. The current programme on ZEUS is a step in this direction.

The irradiation of emulsions is carried out at reduced power for very short periods. Fortunately the ratio of neutron flux to gamma-radiation background inside the core and envelope of ZEPHYR is fairly favourable, so that the amount of fog resulting from this background is not excessive.

The ^{3}He spectrometer has been described by Batchelor, Aves and Skyrme.* It consists essentially of a proportional counter filled with ^{3}He and a heavy stopping gas (krypton). The spectrum of pulse heights from the reaction

$$^{3}He + n \rightarrow {}^{1}H + {}^{3}H + 770\ keV$$

is analysed in a kicksorter and from this and a knowledge of the variation of the cross-section for the reaction with neutron energy it is possible to deduce the energy distribution of the incident neutrons. The method is particularly useful for measuring the energies of monokinetic neutron groups. In a continuous spectrum it is rather difficult to apply, particularly if there are present relatively large quantities of neutrons with energies above 1 MeV, since these can give rise to ^{3}He recoils with energies in the same range as those of the reaction pulses. Corrections for secondary electron pulses due to background gamma radiation and for wall effects also contribute towards the difficulties of analysing continuous spectra. The counters are necessarily rather large to minimize wall effects—those employed in ZEPHYR were $2\frac{1}{4}$ in. outside diameter and $9\frac{3}{8}$ in. long.

* BATCHELOR, AVES and SKYRME. *Rev. Sci. Instrum.*, **26**, p. 1037 (1955).

SESSION 4. EVENING LECTURE

Chairman: SIR JOHN COCKCROFT, K.C.B., C.B.E., D.Sc., F.Inst.P., F.R.S. (*President*)

At this evening session Professor R. E. Peierls, C.B.E., D.Sc., F.R.S., of the University of Birmingham, delivered a lecture on "Recent developments in nuclear theory." In it he referred to the following topics:

the shell model of nuclear structure and its relation to our knowledge of nuclear forces;

collective motion and surface rotations;

highly excited states and the relation between the "optical" picture and the "liquid drop" model;

the size and shape of the nucleus;

high energy radiations as tools for nuclear studies;

prospects of a theory of nuclear forces.

SESSION 5. LECTURE

Chairman: C. SYKES, D.Sc., F.Inst.P., F.R.S.

The effects of irradiation on the physical properties of solids

By A. H. COTTRELL, B.Sc., Ph.D., F.R.S., Atomic Energy Research Establishment, Harwell, Berkshire

Ionizing radiations dissipate most of their energy in solids by exciting electrons. Severe radiation damage is produced in organic substances by this process, but in metals the free electron structure is immune to ionization damage. Atoms are displaced by collisions with fast particles and the point defects so created alter the electrical and thermal properties of solids. These point defects interact strongly with dislocations so that the mechanical properties of metals are sensitive to heavy-particle bombardment. In semi-conductors the displaced atoms can trap electrons and holes, thereby producing large changes in electrical properties. Prolonged bombardments produce drastic effects; some minerals become non-crystalline, uranium crystals change shape, some metals become brittle, and rare gas atoms created by transmutations may produce swelling and disintegration at high temperatures.

RADIATION DAMAGE IN REACTOR MATERIALS

When natural uranium is irradiated in a reactor such as BEPO for a year, about one atom in 7000 undergoes fission. Most of the energy released, about 160 MeV, is carried away as kinetic energy by the two fission fragments. Although it starts at a speed of about 10^9 cm s^{-1} a fission fragment interacts so strongly electrostatically with nearby electrons and nuclei that it is brought to rest in the metal in a distance of only 4×10^{-4} cm.

An energy of 160 MeV is more than sufficient to vaporize 10^7 atoms, or to melt all material in a "thermal spike" 10^4 atoms long and 100 atoms radius. Practically all of the kinetic energy of the fragments is given in the first place to the electrons, and is then transferred to the lattice as a result of electron-lattice collisions. Only about 1% is spent directly in displacing atoms through knock-on collisions between the fragments and atomic nuclei; but this is enough to produce 25000 displaced atoms from each fission, according to Seitz.[1] Every atom in uranium is thus knocked out of place in the lattice a few times during irradiation for a year in a reactor such as BEPO. In power reactors, where the neutron fluxes are higher and the uranium may be enriched with fissile material, radiation damage occurs at correspondingly faster rates.

It is remarkable that the material retains any semblance of its original state in such circumstances. The fact that it does so shows that healing of the crystal structure, back towards its original state, takes place while it is being damaged. This ability to recover rapidly from radiation damage is in fact a crucial property of a reactor material, and much effort has gone into the study of the annealing behaviour of irradiated materials. From this point of view liquids make ideal reactor materials, provided they can be prevented from dissolving their containers. Amongst solids, a metal or semi-metal with a simple lattice of cubic symmetry offers the best prospect. Neither of the two main reactor materials used so far, uranium and graphite, has a lattice of cubic symmetry at the reactor temperature and, in consequence, difficulties have been experienced with both due to changes of shape produced by radiation. The rate of annealing of displaced atoms depends on the rate at which lattice point defects (interstitials and vacancies) migrate through the lattice and hence is very sensitive to temperature. This has led people to make irradiations at low temperatures, down to that of liquid helium, in order to study the point defects before regrouping and recombination begin to take place. Conversely, there is the promise that in more advanced types of reactors working at higher temperatures radiation damage due to displaced atoms will become less of a practical problem. At high temperatures, however, another problem is likely to appear. About 10% of the new atoms created in uranium by fission are the rare gases, krypton and xenon. At temperatures above about 500° C these can diffuse through the lattice to form gas pockets which expand the metal. It is possible that helium, created in certain types of materials suitable for moderators or fuel cans, may produce similar effects. An example of a nuclear reaction from which helium is produced is the $(n, 2n)$ reaction in beryllium:—

$$Be^9 + n \rightarrow 2\,He^4 + 2n$$

EXCITATION OF ELECTRONS BY RADIATION

A fast ion moving through matter loses most of its energy by exciting electrons. The reason for this is that the basic physical quantity transferred in an electrostatic collision is momentum, the amount transferred being determined by the product of the force between the particles and the time of the encounter. For a given momentum p transferred to the "struck" particle, the energy $p^2/2m$ transferred to it varies inversely with its mass. Thus about 1000 times more energy is spent on the electrons than on displacing atomic nuclei. Electronic excitation ceases only when the speed of the ion becomes small compared with that of the most easily ionized electrons in the material. Particles moving through solids of their own kind practically cease to excite electrons when their energies fall below 10^3 to 10^5 eV.

Substances differ sharply in their response to electronic excitation. In metals the existence of a continuous band of energy levels above the Fermi surface enables the excited electrons to drop back easily to this surface, their energy being released eventually to the lattice in the form of heat. There is thus no permanent electronic damage.

In insulators the electrons are excited into the conduction band, which is separated by a finite gap from the valence band. While in the conduction band they can constitute an electric current, which is useful in photoconducting devices and diamond counters, but may raise problems of electrical insulation in nuclear reactors. Thus the electrical resistivity of polyethylene decreases from $10^{17}\,\Omega$ cm to $10^{14}\,\Omega$ cm during irradiation with γ rays at 1000 roentgens per minute

(the γ-ray flux in a reactor is typically of order 10^5 r/min).[2] Mostly, the excited electrons drop back into vacant quantum states ("holes") in the valence band, or get trapped at impurities or lattice imperfections where they form centres for optical absorption, such as the colour centres (F centres) in alkali halides. An interesting branch of solid-state physics has grown up round the study of trapped electrons by optical and paramagnetic resonance absorption measurements.[3]

Drastic molecular changes can be produced in long-chain organic substances by ionizing radiations.[4] The mechanism appears to be that an electron is first excited or ionized somewhere in the chain by the radiation. The excited state is then transferred from bond to bond along the molecule (like an "exciton" in a non-metallic crystal) to a weak bond, usually on a side chain, which is then broken. The free radical that breaks off is chemically active and attacks a nearby long-chain molecule. In some cases this causes neighbouring molecules to become joined together by side chains ("cross-linking"), which is the basis of a useful method for stiffening polythene[5] and vulcanizing rubber. In other cases a main chain is broken, which leads to a reduction in molecular weight and in some cases to a swelling of the substance due to the liberation of gaseous molecular fragments. An example is polymethylmethacrylate.[5] It is interesting to note that polymers that contain benzene rings are much more resistant to radiation damage than others. For example, a dose of 10^{10} r produces little change in the properties of styrene polymers or mineral-filled phenolics, apart from darkening them.[6] It appears that the closed ring of carbon atoms traps the excited electronic state, which then resonates round the ring, without fracturing the molecule, until the electrons fall back into their ground state.

DISPLACEMENT COLLISIONS

Calculations of the number of displacement collisions produced by various charged particles and fast neutrons have been made by several authors.[7] The main points that emerge are as follows:—

(*a*) Owing to the high speed of the incident particle the time of collision with a lattice atom ($\simeq 10^{-17}$ s) is much smaller than the time required to transmit an elastic force from one atom to its neighbours ($\simeq 10^{-13}$ s). The theory of collisions with isolated atoms can thus be carried over to the solid state.

(*b*) The threshold energy E_0 that must be received by the knocked-on atom for it to leave its atomic site is about 25 eV. This value was first estimated by Seitz[1] and has since been confirmed to within a factor of 2 by measurements on several materials. The experimental method is to bombard the material with fast electrons ($\simeq 1$ MeV) and to increase their energy progressively until signs of knock-on damage first appear. The maximum energy transferable to an atom of mass M by an electron of mass m and energy E is given by

$$E_{max} = 2E(E + 2mc^2)/Mc^2$$

where c is the velocity of light. Knowing the minimum experimental value of E for damage, E_0 can then be found by taking $E_0 = E_{max}$. An example of such measurements, obtained on nickel, is shown in Fig. 1.

(*c*) The distribution of damage along the track depends on the displacement cross-section. A fast neutron (e.g. 2 MeV) produces widely spaced collisions (a few cm apart) but at each collision enough energy is transferred (e.g. 10^5 eV) for the knocked-on atom to form a cluster of a few hundred displaced atoms. At the other extreme, an incident electron with energy near the threshold value produces only one displaced atom; the simplicity of the damage in this case has led to the use of electron irradiation in fundamental studies of radiation damage. The behaviour of a charged ion depends on its speed. At very high speeds (e.g. 10^6 eV) it penetrates the innermost electron shell and makes a Rutherford collision with the nucleus. Owing to the high speed of the ion the time of the collision is very small and little energy is transferred unless the nucleus is approached closely. The cross-section for the collision is thus only a few barns and the damage is spread sparsely along the track. As the ion slows down (by loss of energy to the electrons) it reaches a range where it collides with the screening electrons of the target atom and where the collision cross-section is much larger. Hence a large number of collisions occur at the end of the track, giving a dense cluster of displaced atoms, or a "melted region" ("displacement spike").

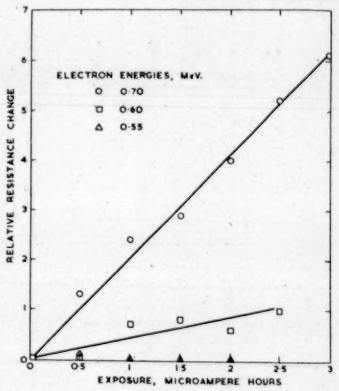

Fig. 1. Change in electrical resistance of nickel foil after exposure to electrons of various energies. Exposure and measurements made at the helium point. Threshold displacement energy is 35($\pm$2) eV. [Work of H. M. Kenworthy, reported in reference (28)]

From the theoretical point of view it is unfortunate that so much of the damage is concentrated at the end of the track, where the calculations are least reliable. Also, being highly clustered, this damage begins to anneal rapidly, and it is difficult to estimate how many of the displaced atoms survive the first few thermal vibrations.

It is thus necessary to develop experimental methods for identifying and counting the displaced atoms. The usual approach is to compare the expected contributions of a vacancy or an interstitial with the observed change in some physical property. A difficulty is that only in the case of one or two properties can the expected contributions be estimated reliably. In all others the comparison is only qualitative, or semi-quantitative. Many of these other properties are of great interest for other reasons, however. We shall now look at some of the main ones. A detailed review has been made by Glen.[8]

NEUTRON SCATTERING

Antal, Weiss and Dienes[9] have developed an interesting method of counting point defects in graphite. A beam of neutrons of initial intensity I_0 is passed in turn through irradiated and unirradiated specimens, and the two transmitted intensities, I_d and I_s respectively, are measured. The neutrons in the beam are sufficiently slow (energy of order 0·001 eV) for their wavelength λ (of order 10 Ångströms) to be too large to produce Bragg scattering in the specimen ($\lambda > 2d$, where d is the largest interplanar spacing for which diffraction is possible), those of higher energy being filtered off. Under these circumstances, if the specimen were perfect the beam would emerge without attenuation. But in practice some of the neutrons are scattered at various defects (e.g. spin defects, thermal scattering, point defects) in the material.

Let σ_0 be the total cross-section per atom for scattering or absorbing neutrons in the unirradiated specimen. Then in a specimen of length x, which contains N atoms per unit volume, the attenuation of the beam is given by

$$I_s = I_0 \exp(-Nx\sigma_0)$$

The advantage of the method lies in the facts that (i) the cross-section for scattering from an interstitial atom is simply the atomic scattering cross-section σ_b which is a known quantity (4·7 barns for graphite), (ii) a vacancy scatters as if it were an interstitial, since it is in effect a negative atom, and (iii) in the irradiated specimen the total scattering cross-section is simply the sum of σ_0 and the total cross-section of the point defect scatterers, $c\sigma_b$, where c is the atomic fraction of point defects. Hence

$$I_d = I_0 \exp[-Nx(\sigma_0 + c\sigma_b)] = I_s \exp(-Nxc\sigma_b)$$

so that c can be determined from the ratio I_d/I_s. Fig. 2 shows the measured neutron intensities transmitted by irradiated ($1{\cdot}1 \times 10^{20}$ n/cm^2) and unirradiated graphite

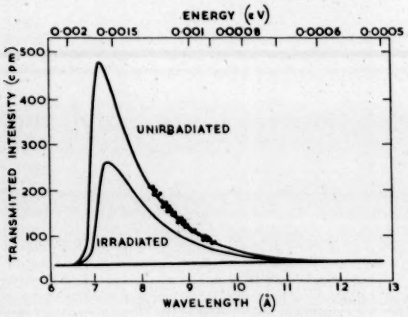

Fig. 2. Intensity of slow neutrons transmitted by an irradiated ($1{\cdot}1 \times 10^{20}$ n/cm^2) and an unirradiated graphite specimen. A typical group of experimental points is shown. Wavelengths shorter than 6·7 Å are cut off by Bragg reflection. (After Dienes[9])

specimens; by measuring areas under the curves the fraction of displaced atoms was found to be 0·0263, which compares well with the value, 0·021, calculated from collision theory.[9]

Applied to materials with small cross-sections for neutron absorption or isotopic spin scattering, this is a promising method for counting point defects, although there may be some ambiguity in specimens where the defects are clustered.

STORED ENERGY

The stored energy method is a fairly reliable one. It is possible to calculate from the theory of interatomic forces the energy stored in a vacancy (≃ 1 eV) and an interstitial (≃ 5 eV), reasonably closely and also to measure the total heat evolved from an irradiated specimen when the defects are annealed. In this way the number of point defects can be determined. For example, Overhauser[10] obtained an energy release of 0·052 calories per gram in copper, after irradiating at the temperature of liquid nitrogen with 9×10^{17} deuterons/cm^2 from a cyclotron. Allowing 5 eV for the energy of an interstitial-vacancy pair, this gives a concentration of 3×10^{-5} displaced atoms.

Fig. 3 shows the total stored energy, and the energy released below 400° C, in graphite after irradiating to various doses at

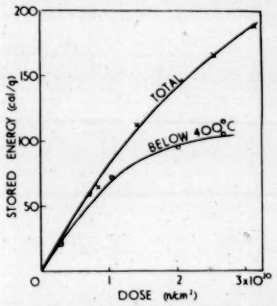

Fig. 3. Total stored energy and energy released in graphite below about 400° C. Irradiation temperature, 30° C. (After Kinchin[11])

30° C.[11] It should be noticed that the energy releases here are large, e.g. 190 cal/g after a total dose of 3×10^{20} n/cm^2, in accord with the large fraction (≃ 1%) of displaced atoms in heavily irradiated graphite.

In heavily damaged materials the heat released on annealing out the damage can become comparable with the thermal energy of the substance. Suppose, for example, that 1% of the atoms are displaced and that 5 eV is stored in each displaced atom. Then the stored energy averaged over all atoms is 0·05 eV, which is equivalent to 0·017 eV per vibrational mode, i.e. to 200° K in temperature. If this energy is released suddenly it may produce a large rise in temperature. This effect was first discovered in 1815 by Berzelius[12] in gadolinite, a radio-active mineral damaged by α particles released by its own activity, in which a release of stored energy sets in so rapidly on heating that the temperature runs away to a white heat.

ELECTRICAL PROPERTIES

Electrical resistivity appears to be an ideal property to use for studying radiation damage in pure metals since it is sensitive and easily measured. Unfortunately, however, it is difficult to estimate reliably the cross-section of point defects

for scattering conduction electrons. Jongenburger[13] (see also Blatt[14]) has calculated that 1% of vacancies raises the resistivity of copper by $1{\cdot}3 \times 10^{-6}$ ohm cm. For comparison, Overhauser[10] observed experimentally in the annealing of irradiated copper that a stored energy of 1·7 cal/g was released for each $10^{-6}\,\Omega$ cm of resistivity annealed out. Ascribing this annealing to the recombination of vacancies and interstitials, and allowing 5 eV per pair, one obtains $11 \times 10^{-6}\,\Omega$ cm per 1% of pairs. This suggests that 1% of interstitials raises the resistivity by $10 \times 10^{-6}\,\Omega$ cm. Earlier theoretical estimates of the resistivity of interstitials gave a much lower value than this, and it appeared that a real discrepancy existed between the resistivity and stored energy measurements. Recently, however, Overhauser and Gorman[15] have calculated the resistivity of interstitials to be $10{\cdot}5 \times 10^{-6}\,\Omega$ cm per 1%; the main contribution is provided by the strain fields round the defects.

A striking discrepancy between stored energy and resistivity measurements has been reported recently by Blewitt, Coltman, Noggle and Holmes.[16] They irradiated copper in a helium cryostat in a reactor and then annealed it through the range of 40° K, where about 40% of the resistivity change annealed out. From the slope of the temperature/time curve over this range, however, it appeared that less than 5×10^{16} displacements/mole annealed out, compared with the 10^{19} displacements/mole deduced from the resistivity measurements. It may be that one is dealing here with the clustering rather than the annihilation of defects, and that the resistivity is much more sensitive to clustering than is the stored energy.

Evidence for grouping of defects is provided by the complex behaviour observed on annealing irradiated metals at low temperatures.[17] An example is shown in Fig. 4. It was

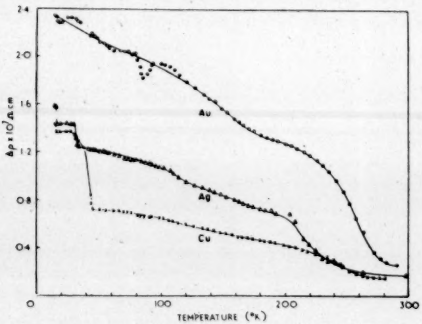

Fig. 4. Annealing of copper specimens after irradiation at 12° K. The residual resistivity after annealing at temperature T is shown as a function of T. (After Cooper, Koehler and Marx[17])

hoped originally that such experiments would quickly yield measured values of the activation energies for the movement of vacancies and interstitials in solids, but the unexpected complexity of the observed behaviour has prevented this, and alternative schemes for interpreting the data now exist in the literature.[8]

One of the most striking results from this work is the demonstration of the high mobility of some types of defects in metals at very low temperatures. Undoubtedly, this ability to anneal out radiation damage rapidly is to the advantage of metals as reactor materials. By comparison, much higher annealing temperatures are necessary in covalent solids. Fig. 5 shows the annealing ranges in graphite.[11]

The complexity of the annealing phenomena in neutron-irradiated materials can be traced partly to the highly-

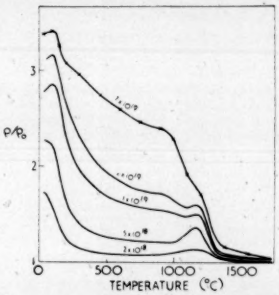

Fig. 5. Recovery of resistivity of irradiated graphite after annealing at various temperatures. The neutron dose is shown on the curves. (After Kinchin[11])

clustered distribution of damage that is produced by knock-on collisions. It is generally agreed that a better way to obtain systematic information about the behaviour of elementary vacancies and interstitials is to resort to other types of experiments, e.g. quenching from high temperature to introduce vacancies, and electron irradiation at low temperature to introduce simple vacancy-interstitial pairs.

The electrical properties of semi-metals and impurity semiconductors are of course highly sensitive to knock-on damage, because the interstitials and vacancies form traps for electrons and holes. Changes in the Hall coefficient have been used to study the early stages of damage in graphite.[11] Germanium transistor devices are quickly ruined in a reactor, an irradiation of a few seconds only ($\simeq 10^{13}$ n/cm^2) being sufficient to change n-type germanium to p-type[18].

THERMAL CONDUCTIVITY

Not much work has been done on this property. An irradiation of 3×10^{20} n/cm^2 at 30° C lowers the thermal conductivity of graphite by a factor of 27; the same dose at 150° C lowers it by a factor of 7.[11] We expect small changes in thermal conductivity in metals, in which the electrons carry the heat. In insulating crystals on the other hand the thermal conductivity decreases strongly, due to the scattering of lattice thermal waves by defects.[19] This may be important in brittle, ceramic, reactor materials since it leads to steeper temperature gradients and larger thermal stresses.

DIFFUSION

In principle, an obvious way to demonstrate the formation of vacancies and interstitials by irradiation is to measure the change in the diffusion coefficient. In practice most experiments of this type have been unsuccessful because radiation damage anneals so rapidly at the high temperatures necessary for large-scale diffusion. Much greater sensitivity is obtained,

however, by measuring the state of order in alloys such as Cu_3Au since the configuration of atoms is altered along the whole track of a migrating vacancy. Thus, Dugdale and Green[20] have shown that disordered Cu_3Au normally does not order at an appreciable rate at temperatures below 170° C, whereas after irradiation it orders readily at 100° C.

X-RAY EFFECTS

In cubic metals changes of lattice parameter are very small and not easily interpretable. In other materials larger changes are produced. For example, in graphite the interstitial atoms produced by irradiation collect in the spaces between the close-packed sheets of carbon atoms and cause an expansion along the *c* axis, perpendicular to the sheets; the *c* spacing increases almost linearly at the rate of 1·3% per 10^{20} neutrons per cm^2 at 30° C.[11] Striking changes are produced in quartz (see Fig. 6), which can lead to volume

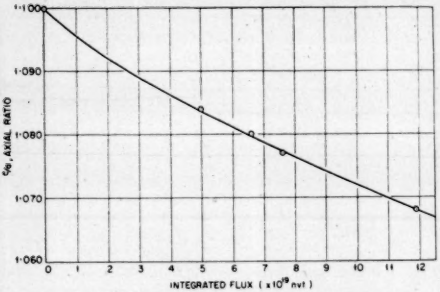

Fig. 6. Change in axial ratio of quartz after irradiation with fast neutrons. (After Crawford and Wittels[21])

increases of 15% without the appearance of any cracks or bubbles, and the crystal transforms to an optically-isotropic glassy state at doses of about 10^{20} n/cm^2.[21] This property of retaining so much displacement damage that the lattice breaks down to an amorphous structure is largely characteristic of silica and silicates, and occurs notably in the "metamict" group of radioactive minerals.[7]

DIMENSIONAL CHANGES

Irradiation causes a bulk growth in polycrystalline graphite ("Wigner growth") which closely follows the X-ray change, but is smaller because the dimensional change is partly accommodated by the pores in the structure. At 30° C the bulk growth perpendicular to the extrusion direction is about 0·1% per 10^{20} n/cm^2.[11]

A very striking growth occurs when uranium is irradiated at temperatures below 450° C. When a single crystal of orthorhombic uranium is irradiated for a year in BEPO at, say, 200° C, it elongates by about 12% along the *b* axis, shrinks by the same amount along the *a* axis, and remains unchanged along the *c* axis.[22] There is no appreciable change in the lattice structure or dimensions so that the effect is due to the movement of atoms from one part of the crystal to another. Wyatt and Pugh[22] developed the view that this movement occurs by plastic deformation in the expanded zones round the thermal spikes along the tracks of fission fragments. An essential part in this theory was played by twinning modes of deformation; this leads to the prediction, which has been confirmed experimentally, that growth ought not to occur at temperatures above about 450° C because uranium deforms plastically by slip instead of twinning at such temperatures. An alternative theory has been put forward[22,23] in which the interstitials migrate to form new sheets of atoms perpendicular to the *b* axis and the vacancies either migrate along the *a* axis to crystal boundaries or are absorbed on dislocations with Burgers vectors lying along the *a* axis. This theory can explain why the rate of radiation growth becomes small at low temperatures,[23] but it is not obvious why the interstitials and vacancies should behave in the highly selective manner that the theory requires.

The large dimensional changes produced by radiation growth are of course quite intolerable in a power reactor, and some way has had to be found to overcome them. Fortunately, the crystallographic nature of the effect suggested a direct solution. By suitable thermal treatment uranium fuel rods can be prepared in the form of fine grained polycrystals with random crystal orientations. Although individual crystals grow, the overall effect, averaged over all orientations, is zero.

The fact that the individual crystals still grow, in a random polycrystal, leads to a further effect which has been studied recently.[24] As each crystal grows it impinges on its neighbours. At first these local growth strains between the crystals can be accommodated by purely elastic deformations, but eventually they become too large and the grains are then forced to deform plastically in order to remain joined together. We thus have the unusual situation of a solid which is continually undergoing spontaneous plastic deformation without external forces applied to it and without changing its external shape. Perhaps the nearest analogy is that of a liquid in which the molecules are spontaneously and continually changing position without external stimulation. How does such a material respond to an external force? Continuing the analogy, we expect it to deform like a liquid of high viscosity rather than like a solid, i.e. we expect it to possess no elastic limit and to deform steadily at a rate proportional (or nearly so) to the applied stress. The rate $\dot{\epsilon}_e$ of this flow is estimated to be

$$\dot{\epsilon}_e = (\sigma/\sigma_y)\dot{\epsilon}_g$$

where $\dot{\epsilon}_g$ is the rate of radiation growth in a uranium crystal, σ is the applied stress and σ_y is the yield stress in the absence of irradiation. Typically $\dot{\epsilon}_g \simeq 4 \times 10^{-9}$ per sec and $\sigma_y \simeq 5 \times 10^4$ pounds per sq in. Hence to produce viscous flow at a rate of 10^{-11}/s an applied stress of order 100 lb/$in.^2$ is required. Such a stress is small enough to be supplied by the weight of the specimen itself. Recent experiments on the creep of uranium springs in BEPO have confirmed that flow occurs at rates given by the above formula.[24]

MECHANICAL PROPERTIES OF METALS

The radiation-creep effect described above is exceptional, due to the unusual growth effects in uranium. In most metals the effect of irradiation is to produce a hardening. Fig. 7 shows an example of the type of change that is produced in the stress-strain curve.[25] The development of a sharp yield point, and various other features, suggest that point defects behave like impurity atoms in their hardening effect. In particular, they appear to migrate to dislocations and to anchor them in position.

A striking demonstration of this is provided by experiments on internal friction.[26,27] If a single crystal of, say, pure copper is vibrated mechanically at room temperature at a frequency of order 10^4 c/s and a strain amplitude of order 10^{-5}, most of the vibrational energy is dissipated through the

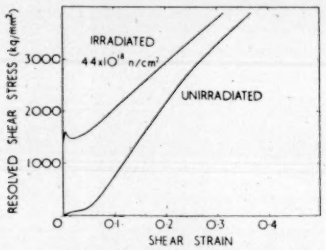

Fig. 7. Effect of neutron irradiation on the stress-strain curve of copper single crystals at room temperature. (After Makin[25])

oscillations of dislocation lines in the crystal. If the dislocation lines are then pinned down in some way this internal friction disappears. It has long been known that extremely small amounts of impurity can effectively suppress this type of internal friction by the segregation of impurity atoms in dislocations. The striking result is that the same effect can be produced by an extremely short irradiation. Thus a few minutes ($\simeq 10^{14}$ n/cm^2) in the reactor is sufficient to reduce the internal friction by an order of magnitude (see Fig. 8). The number of displaced atoms produced by an irradiation

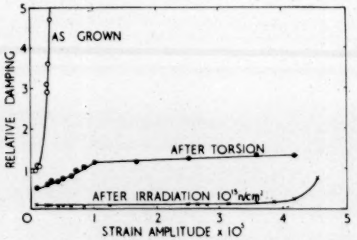

Fig. 8. Internal friction at room temperature of a copper single crystal measured (*a*) as grown, (*b*) after torsion, (*c*) after neutron irradiation. (After Barnes and Hancock[27])

as short as this is extremely small, not more than about 1 in 10^7. This is undoubtedly the most sensitive technique we have for detecting the earliest stages of radiation damage in pure metals.

Heavy irradiations produce strong hardening and some metals even become brittle. This is particularly true of those body-centred cubic metals (e.g. iron and molybdenum) that are normally brittle at low temperatures; radiation produces a marked increase in the ductile/brittle transition temperature. For example:—

Material	*Irradiation treatment*	*Transition temperature Before irradiation*	*After irradiation*
SA70 steel[28]	$2{\cdot}5 \times 10^{19}$ n/cm^2 at 50° C	−30° C	+50° C
Molybdenum[29]	10^{20} n/cm^2 at 80° C	−30° C	+70° C

Uranium itself also becomes brittle at room temperature after an irradiation of about 10^{19} thermal neutrons/cm^2.[23]

ACKNOWLEDGEMENTS

I am grateful to Dr. M. J. Makin and Mr. R. S. Barnes for allowing me to show unpublished results of theirs, in Figs. 7 and 8.

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DISCUSSION ON SESSION 5

Mr. W. Simpson: In his remarks on the effect of irradiation on high molecular weight organic compounds, Dr. Cottrell has mentioned that the electronic excitation is transferred along the molecule to a weak bond which is then broken, and that this weak bond is usually on a side chain. This may be demonstrated fairly readily. In order to study the scission of branched hydrocarbon polymers, we have carried out experiments on linear polymethylene and several lightly branched polymethylenes containing known side chains. The gaseous hydrocarbons given off by these polymers on irradiation were analysed mass-spectrometrically.

The results for the linear polymethylene are shown in Fig. 1, and if we compare this with the results for the branched polymers, it can be seen that there is a selective removal of the short side chains (Figs. 2, 3 and 4). The side chains appear in the gaseous product as saturated hydrocarbons. Methane is the only other hydrocarbon produced in comparable yield.

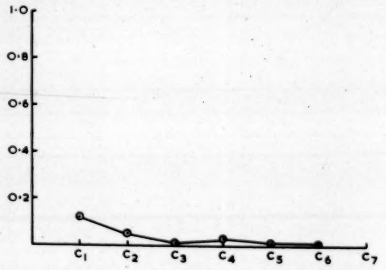


Fig. 1. Polymethylene

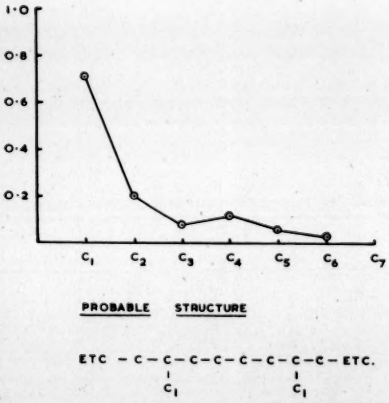


Fig. 2. Methyl branched polymethylene

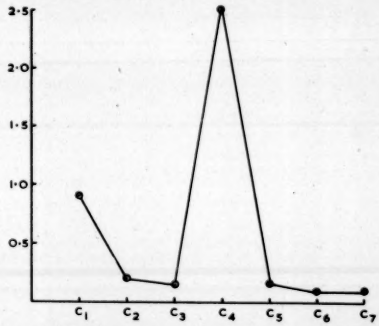


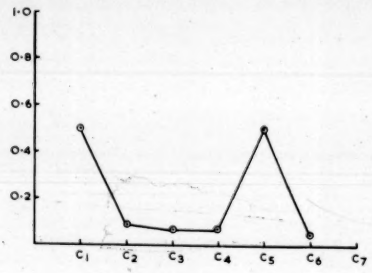
Fig. 3. Butyl branched polymethylene

Unsaturated hydrocarbons are also produced but these are not regarded as important in the present context.

This form of radiation damage is extremely useful because it yields information not obtainable by other means. Infrared spectroscopy can give an estimate of the number of branches on an unknown hydrocarbon, but so far gives little information on their length. In so far as the analysis of the saturated hydrocarbon gases evolved on irradiation supplies this information directly, it is potentially an important new technique in polymer examination.

An example of this is given in Fig. 5 in which the gases evolved by two polyethylenes are compared. The polymer prepared by high pressure synthesis appears to contain mainly ethyl and butyl side chains. Very long side chains

cannot of course be detected by this technique. Even if recombination of the broken primary link to the main chain does not occur, the hydrocarbons are probably too involatile to be detected, although this point is being checked. It is also important to note that it is evident from this experiment that



PROBABLE STRUCTURE.

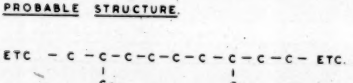


Fig. 4. Amyl branched polymethylene

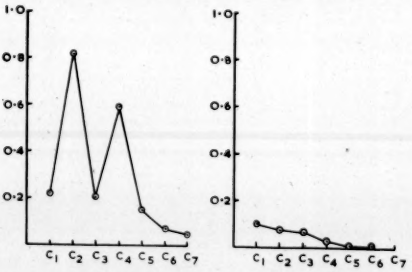


Fig. 5. Comparison of polythenes

(*a*) high pressure polythene: density 0·920 g/c³, B.S. softening point ~60° C.
(*b*) low pressure polythene, density 0·970 g/c³, B.S. softening point ~110–120° C.

methane is not a necessary scission product of branched hydrocarbons. This suggests that the branched polymethylenes may all contain methyl side chains due to the operation of some unsuspected mechanism during their preparation.

The hydrocarbons evolved from the low pressure polyethylene on the other hand are very similar in their amount and distribution to those given off by the linear polymethylene, and show that the polymer is considerably more linear than the high pressure polymer. This evidence is in line with other physical measurements on these polymers. The closer chain packing possible with linear molecules is reflected in the higher density and softening point of the low pressure polyethylene.

Mr. C. G. Garton: Measurements of intrinsic electric strength of β-irradiated polythene which I propose to describe illustrate two points; one, that high-energy irradiation may be a valuable way of changing one property of a material for the purpose of elucidating the theory of that property, while leaving the general nature of the material unchanged; secondly, that one should not assume without investigation that any observed change is necessarily a direct result of irradiation. It may, as in the present case, be a secondary result of a change in another property.

The instrinsic electric strength of a solid dielectric is the strength observed when all secondary causes of failure, such as ionic bombardment and thermal instability, have been eliminated by suitable experimental techniques. It occurs when the field strength is sufficient to impart energy to the electrons of the solid more rapidly than they can transfer energy to the solid structure.

The instrinsic strength of normal polythene was measured some fifteen years ago, and has been confirmed since, with results shown by curve *C* of Fig. 6. The marked fall of

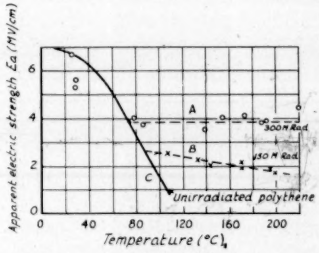


Fig. 6. Variation of electric strength of polythene with temperature. (Values calculated from the initial sample thickness)

Curves A and B. Effect of 4 MeV radiation.
Curve C. unirradiated specimen.

strength with increasing temperature was predicted theoretically, although the temperature at which the fall should occur could not be calculated. It was supposed that the observed fall reflected the theoretical result.

On the basis of the same theory, when the possibility arose of comparing results with irradiated polythene, it was expected that the electric strength would be either unaffected by irradiation, or would be increased at low temperatures and decreased at high ones, resulting in an even sharper fall.

The actual results, shown for two degrees of irradiation by curves *A* and *B* of Fig. 6, were wholly unexpected. The strength fell exactly as for the unirradiated material up to a certain temperature, but then diverged sharply from the original results and remained practically constant over the whole of the remaining temperature range. This was unintelligible on the existing theory.

It was then observed that measurements of the modulus of elasticity of irradiated polythene (by Charlesby and Hancock,[1] see Fig. 7) showed very similar characteristics, the lowest two curves of the figure corresponding in degree of irradiation to curves *A* and *B* of Fig. 6.

This suggested that breakdown above 20° C was not, in fact, intrinsic, and not a purely electrical phenomenon at all, but was due to some form of mechanical failure under

electrostatic forces. Analysis of the equilibrium of a plane slab between free plane electrodes shows that a mechanical instability can indeed exist. Contrary to most mechanical systems, in which deformation either relieves the load or

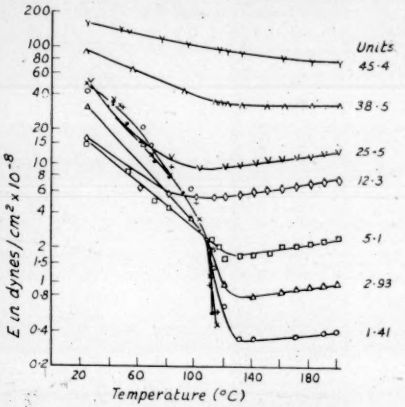


Fig. 7. Variation of modulus of elasticity with temperature and radiation dose (dynamic method). (After Charlesby and Hancock[1])

• 0
× 0·36
+ 0·62

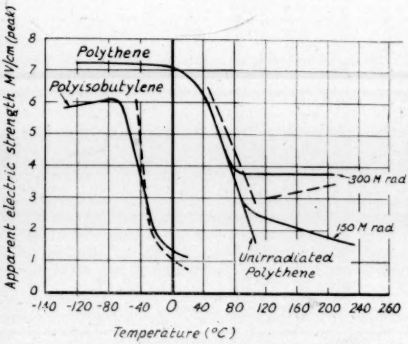


Fig. 8. Electric strength limited by mechanical failure. (After Stark and Garton[2])

——— experimental values
– – – – calculated maximum electric strength from Young's Modulus data

leaves it unaltered, a dielectric slab between free electrodes at constant potential is subjected to a larger force the more it yields. It is easy to show[2] that stable compression to less than 60% (actually $e^{-\frac{1}{2}}$) of the initial thickness is impossible, a d that any higher electrical stress leads to an instability, with complete collapse of the slab. The electrical stress, in e.s.u./cm, at which this occurs for a plane slab is given by

$$E_y = (4\pi Y/\varepsilon)^{\frac{1}{2}}$$

where Y and ε are the modulus of elasticity and the permittivity respectively.

The irradiated material gives the abnormal curves A and B in Fig. 6 simply because the value of Y in the above equation behaves similarly, as in Fig. 7. The cause of the abnormal behaviour of the modulus of elasticity is, of course, that the material which has been cross-linked by irradiation possesses a rubber-like, or entropy-elasticity, in addition to its normal elasticity, and at high temperatures the former predominates, with a very different type of temperature dependence.

Fig. 8 compares the experimental results with calculated curves based on the hypothesis of mechanical failure, and shows good agreement not only for the irradiated polythene but also for normal polyisobutylene, for which independent mechanical and electrical data happened to be available.

Dr. A. Charlesby: The previous two speakers have given instances where radiation-induced changes may be useful as an analytical tool. I would like to discuss several items where basic problems involved in radiation-induced changes remain to be solved.

In carrying out research on the effects of radiation on materials, one is immediately struck by the enormous difference in their sensitivity to radiation. Most metals may be used in very intense fields, such as inside atomic reactors or as anodes on X-ray machines for periods of the order of years without marked changes in their physical properties under usual operating conditions. Under similar conditions, ordinary organic materials are permanently modified after a period, usually of days, whereas plastic materials suffer considerable changes after atomic pile radiation of the order of one hour or less.

The major effect arises in metals from the displacement of atoms. This is accompanied by intense local heating caused by the enormous energy dissipated locally. One may then envisage the subsequent changes as depending on two factors—the rate at which this heat is lost and the time taken for atoms to move to new and stable positions in the lattice. If the rate of heat loss from the site of the initial interaction is more rapid than the re-adjustment of the atoms, the changes produced should be, in many respects, comparable with quenching. On the other hand, if the rate of dissipation of heat is lower than the rate of re-arrangement of atoms, displaced atoms will have sufficient time and certainly sufficient energy to move to more stable sites and the net effect of radiation will be more analogous to annealing.

The most profound changes occur in irradiated organic materials and here several fundamental problems arise whose solution may provide interesting information on the basic nature of reactions in solids.

In the first place, the effect of radiation on such materials is to produce ions, excited atoms and free electrons. This process is reasonably well understood. However, subsequent examination of the products showed that these can best be explained by reactions between radicals, i.e. uncharged molecules with an unpaired electron. There are, therefore, at least three stages in the complete reaction:

(i) a physical one resulting in ionization and excitation
(ii) the transformation of these to radicals
(iii) the interaction of the resultant radicals.

The major theoretical difficulty is to explain quantitatively the transformation from the ionized or excited molecules to

the radicals. A number of theories have been advanced but the position is far from clear.

The second problem arises from energy transfer. The initial phenomena of ionization or excitation take place at random throughout the system while the final chemical changes can be very specific in character. One must therefore postulate a mechanism whereby the initial effect may be transferred along the molecular chain or even from one molecule to a neighbour. This problem of energy transfer, which is fundamental in radiobiology is becoming of increasing importance in the study of radiation effects and even in atomic pile design, where it may influence the types of materials which can be used.

The third, and probably related problem, concerns the so-called temperature effect. The energies required to produce many chemical changes are close to those required to produce an ion pair in air, i.e. about 35 eV. However, it is found that many of these reactions produced by radiation depend very much on the temperature. This is very surprising since the energies involved in these chemical changes, being of the order of tens of electron-volts, would correspond to energy differences equivalent to many thousands of degrees, while the temperature dependence observed shows that the resultant changes can be modified by a thermal energy change corresponding to 0·01 eV or less. Here again, one must visualize some form of system activated by an initial ionization or excitation which lasts sufficiently long to enter into thermal equilibrium with its surroundings before the final reaction to a straightforward chemical change takes place. This time scale must again be closely related to that existing in metals.

The fourth problem concerns the comparison of chemical changes produced in photochemistry with those arising in high energy radiation. Ultra-violet light of suitable wavelength can cause excitation of specific electrons depending on resonance effects whereas high energy radiation will be much more indiscriminate. It is of considerable importance to compare in detail the nature of the chemical changes produced by these two forms of radiation.

If I may conclude on a rather different note, I would like to raise the question of terminology. In most of this work, we classify together X-rays, gamma rays, fast electrons and energetic atomic particles under the name of "ionizing radiation" since these radiations are able to produce ionization. On the other hand, ultra-violet light will generally only produce excitation and should, by analogy, therefore be termed "exciting radiation." In practice most phenomena observed with high energy radiation can be explained in terms of either displaced atoms or excitation, and ionization is not necessarily the most important effect produced. It may therefore be preferable to refer to "atomic radiation" as any form of radiation which can produce atomic re-arrangements, either in the electronic structure or in the position of the nucleus, and to "nuclear radiation" when dealing with radiations capable of causing changes in the nucleus itself.

Dr. J. Spaepen: Is the use of convertors in a thermal reactor likely to increase the contribution of fast neutrons to the irradiation damage? Can the time, necessary to obtain a given effect, be reduced considerably?

Dr. J. B. Birks: I wish to refer briefly to one aspect of radiation damage in solids that has received relatively little attention, namely the decrease in the fluorescence or scintillation efficiency of crystal phosphors produced by irradiation.

Birks and Black[3] studied the deterioration of crystalline anthracene due to irradiation by ^{210}Po α-particles, and found that the relative scintillation efficiency decreases according to the simple relation

$$S/S_0 = (1 + AN)^{-1}$$

where N is the total number of particles/cm^2 striking the crystal. A is a parameter, characteristic of the material and the particular type of irradiation. Its reciprocal A^{-1} represents the value of N required to reduce the scintillation efficiency to half its original value. Black[4] extended this investigation to several other organic crystals, and found the above relation to apply also to these. The values of A^{-1} for the different materials irradiated by 3 MeV α-particles are listed in Table 1. Irradiation with up to 10^{13} electrons/cm^2 of

Table 1. *Values of* A^{-1} *for materials irradiated by 3 MeV α-particles*

Anthracene	*Naphthalene*	*Terphenyl*	*Stilbene*	*Diphenyl acetylene*
5×10^{10}	$6{\cdot}7 \times 10^{10}$	$2{\cdot}1 \times 10^{11}$	$2{\cdot}7 \times 10^{11}$	$1{\cdot}25 \times 10^{12}$

average energy 40 keV, produced no measurable decrease in scintillation efficiency, showing that the effect is due specifically to heavy particles.

Similar measurements on various inorganic phosphors have been made by Marsden,[6] Broser and Kallmann[5] and others. Although these observers omit to note this, an analysis of their results indicates that the relation quoted is also valid for inorganic phosphors. The derived values of A^{-1} for ^{210}Po α-particles are listed in Table 2. The similar values for the copper and silver activated ZnS phosphors indicate that the damage occurs in the crystal lattice, rather than in the impurity activator centres, and is probably due generally to the formation of "quenching" centres.

Table 2. *Derived values of* A^{-1} *for* ^{210}*Po α-particles*

CdS	*ZnS (Cu)*	*ZnS (Ag)*	*Zn_2SiO_4*	*$CaWO_4$*
$2{\cdot}5 \times 10^{12}$	$2{\cdot}5 \times 10^{13}$	$2{\cdot}5 \times 10^{13}$	$1{\cdot}3 \times 10^{14}$	∞

From radiation-chemical studies, it is known that aromatic compounds, of which the materials listed in Table 1 are examples, are much less prone to radiation damage than aliphatic compounds, probably due to the capacity of the π-electron system to absorb and store excitation energy. Table 1 shows that the condensed hydrocarbons, anthracene and naphthalene, are however more susceptible than the other three hydrocarbons, containing phenyl rings. The inorganic phosphors are seen to be less liable to radiation damage than the organic phosphors, due probably to their higher valence-bonding energies, and higher atomic masses. Calcium tungstate is remarkable in that no observable decrease in scintillation efficiency occurs on prolonged α-particle irradiation.

Apart from the scientific interest of these effects, they also have technological significance for nuclear reactor instrumentation. Tiny scintillation crystal units, located within the reactor core, and linked through suitable light guides to external photomultipliers, represent an attractive alternative to the rather cumbersome ionization chambers and counters currently used for reactor measurements.

May I ask Dr. Cottrell whether any radiation damage studies have been made at Harwell or elsewhere of crystal phosphors under neutron irradiation, with a view to their utilization as scintillation detectors within a reactor?

Dr. H. S. Arms: Perhaps Dr. Cottrell would be so good as to explain whether there is any physical significance to the Z-shape portion of the curve in his paper. This shows the elongation of the spring as continuing in a straight line in two

instances independent of whether or not the spring is in a flux field. Or is the information which he has accurate enough to settle this problem at all?

Mr. K. Thomas: With reference to the permanent damage of irradiated materials, I would like to ask Dr. Cottrell why our attention is particularly directed to changes in dislocations as the possible explanation, and not to other causes? And further, whether the investigation of this damage has advanced to a stage where he could expound on the changes envisaged?

Dr. W. Humbach: Dr. Cottrell showed several slides pertinent to neutron irradiation, but only one pertinent to deuteron irradiation. Since the latter method, as I understand it, is a very fast one, which is of great practical importance, I would like to ask Dr. Cottrell whether he would make some comments on the relevance of the deuteron data and the limits of the method.

Mr. J. E. B. Perkins: Dr. Cottrell mentioned that uranium metal was found to be softer than normal while under irradiation, but harder when investigated after removal from the reactor. Is this effect to be expected in a lesser degree with other metals such as steel? If so, then a considerable amount of published information giving physical properties of materials, as measured after irradiation, may be entirely misleading for reactor design purposes.

Another problem, which is of great interest to the reactor engineer, is that of metals in close contact and under irradiation. Metals may be required in static or in moving contact, or alternatively in a system involving close clearances.

It has been suggested that there are two distinct problems: (*a*) owing to the growth of the materials under irradiation, excessive loading or seizure may occur; (*b*) owing to fast neutron reactions causing "hot spots" in the materials, there may be local welding effects at the surface. Would Dr. Cottrell comment on any experimental evidence he has available of such effects, or failing this give his opinion as to whether he would expect such effects to be serious from the more fundamental considerations?

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AUTHOR'S REPLY

Dr. A. H. Cottrell: The answer to Dr. Spaepen's question is that convertors can and are being used in thermal reactors to increase the contribution of fast neutrons in irradiation experiments. The saving of irradiation time obtained with their use appears, however, to be smaller than at first anticipated; this may perhaps be due to annealing of disorder within the large thermal spikes produced by fast neutron collisions.

In reply to Dr. Birks, I believe that Dr. Garlick is interested in effects of neutron irradiation on crystal phosphors.

We are very intrigued by the Z-shaped portion of the uranium creep curve referred to by Dr. Arms, but do not have enough experimental evidence yet to assess its physical significance. Certainly, I would expect the creep deformation to continue for a period after the neutron flux has ceased, since the internal stresses that cause the creep do not disappear directly with the removal of the flux. The subsequent reversal of the strain is unexpected, and we are now making further experiments to investigate it.

In reply to Mr. Thomas, the reason why I think changes in dislocations may be the cause of radiation hardening is that this hardening persists to temperatures well above those at which vacancies and interstitial atoms are known to anneal out. It is too early to be dogmatic on this point, since the question of whether stable clusters of vacancies or interstitials can be formed has not been settled, but I think that the weight of the evidence points to some form of damage to the dislocation lines themselves as the cause of the hardening. The suppression of mechanical damping (i.e. dislocation oscillations) by irradiation strongly indicates such an effect. Perhaps what happens is that the point defects are absorbed onto dislocation lines with the result that the latter no longer lie in good slip planes and in consequence their glide motion becomes resisted by large Peierls forces.

I agree with Dr. Humbach that deuterons are very useful for irradiation experiments. A characteristic feature of deuteron irradiation is that high-energy particles (e.g. 10 to 20 MeV) have to be used in order to achieve a useful depth of penetration in the specimen, in which case the energy delivered to the lattice in any single collision is rather small, i.e. a few hundred electron volts. These collisions thus differ appreciably from those with fast neutrons, in which large thermal spikes are produced, and it would be risky to extrapolate from the effects of deuterons to those of neutrons. On the other hand, the small spikes produced by deuterons are a little easier to analyse theoretically than those of neutrons, so that in purely fundamental studies of radiation effects there is some advantage in using deuterons, quite apart from experimental considerations.

In reply to Mr. Perkins, I think it is doubtful whether materials other than uranium will appear appreciably softer during irradiation, since the effect in uranium depends on its unique radiation-growth behaviour. Perhaps a creep-resistant alloy, working near the upper limit of its useful temperature range, may show an increased creep rate due to the creation of point defects by the radiation, but I know of no evidence at present that such an effect exists. The question of the frictional behaviour of metals under irradiation is very interesting, but I do not know of any experiments. I doubt whether the hot spots produced from neutron collisions would add significantly to the effects of those already produced in sliding friction. Certainly, if the material grows under irradiation seizure is likely to occur.

SESSION 6. DISCUSSION ON "ANCILLARY PROBLEMS OF REACTORS"

Chairman: H. S. ARMS, D.Phil., F.Inst.P.

Shielding for reactors

By G. C. LAURENCE, M.B.E., Ph.D., F.R.S.C., Atomic Energy of Canada Ltd., Chalk River, Ontario, Canada

A brief introductory discussion is given of the design of shielding for nuclear reactors. It includes consideration of design requirements, choice of materials and dimensions, nuclear reactions that affect the penetration, heating of the shielding and production of radioactivity in the coolant. Because the basic data available are insufficient for high accuracy in design calculation, the use of very complex theory and very laborious calculation has little advantage. Very approximate methods are used, and allowance made for possible error in calculating the thickness of shielding required. The problem is simplified by neglecting radiation processes that have relatively little effect on the overall penetration, but the importance of these effects depends on the design of the shielding.

The most widely recognized authority on radiation hazards is the International Commission on Radiological Protection. Its recommendations, as revised December 1, 1954, were published by the British Institute of Radiology,[1] and in other radiological journals. The Commission recommends for those who are exposed to penetrating radiation in their normal employment that the dose of gamma radiation absorbed in any part of the body should not exceed 30 ergs/cm^3 in any week. They recommend that the energy absorbed from fast protons produced by collisions of fast neutrons should not exceed 3 ergs/cm^3 per week.

In the design of reactor shielding to conform to these recommendations, the intensities in the shielding near its outer boundary next to areas occupied by personnel during normal working hours should not exceed the following limits:

7·5	mr/h of gamma rays
2000	thermal neutrons cm^{-2} s^{-1} flux
30	fast neutrons cm^{-2} s^{-1} flux

The dosage rate produced by one photon cm^{-2} s^{-1} is given in Fig. 1 as a function of its quantum energy.

The passage of gamma rays and neutrons through the shielding depends in a complex manner on the energies of

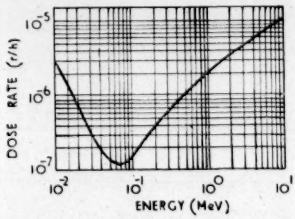

Fig. 1. Gamma ray dosage rate produced by 1 photon cm^{-2} s^{-1}

the rays and particles, the kinds of the nuclides they encounter and the shape and arrangement of the shield. Precise calculation of the penetration is mathematically difficult and laborious. The use of accurate detailed theory is at present scarcely warranted because it does not yield the accuracy desired owing to the inaccuracy of some of the data that is required. In practice much simpler and more approximate methods are commonly used, and the shielding thicknesses are increased to allow for possible error.

In this brief introduction to reactor shielding it is not possible to discuss the theory of penetration of radiation except to describe in simple non-mathematic terms the more important effects. Comprehensive theory is given in a number of well known texts, and its application to shielding design is contained in the *Reactor Shielding Manual*.[2] A helpful source of information for those having access to classified documents is a report by Henderson and Whittier.[3]

The primary source of radiation in the fuel may be described approximately as follows: The release of 1 W of power by nuclear fission is accompanied by

$7{\cdot}8 \times 10^{10}$ n/s
$5{\cdot}2 \times 10^{10}$ penetrating photons/s

The number $N(E)$ of neutrons emitted per watt with energy between E and $E + dE$ is given approximately by

$$N(E) = 7{\cdot}8 \times 10^{10} \times \sqrt{\frac{2}{\pi e}} \sinh \sqrt{(E/0{\cdot}5 \text{ MeV})} e^{-E/1 \text{ MeV}} dE \quad (1)$$

for energies up to 17 MeV. The gamma rays have widely distributed energies but may be grouped to simplify calculations as

5×10^{10} photons/s of average energy 1·36 MeV
2×10^{9} photons/s of average energy 5 MeV

To this should be added the gamma rays produced by radio-active neutron capture in the fuel. The important penetrating components emitted with 1 W of fission power may be grouped for natural uranium as follows

$2{\cdot}5 \times 10^{10}$ photons/s of average energy 0·8 MeV from fission products
10^{7} photons/s of average energy 2·8 MeV from fission products
3×10^{10} photons/s of average energy 5 MeV from fissile and fertile nuclides.

The important penetrating gamma rays produced by neutron capture in other materials commonly used in the reactor core are given in Table 1.

Table 1. *Penetrating gamma rays from neutron capture in reactor materials*

Capturing element	*Capture cross-section*	*Number of photons per neutron captured*	*Approximate energy of photon* (MeV)
Hydrogen	0·336	1	2·2
Aluminium	0·21	0·35	7·7
		0·8	4
		1	1·8
Iron	2·43	0·5	8
		0·2	6
		0·2	4
Lead	0·17	0·93	7·4
		0·07	6·7
Zirconium	0·18	1·1	4
		0·4	6
		0·1	7·5
Beryllium	9·0	0·75	6·8
		0·5	4
Sodium	0·47	1	2·8
		1	1·4
		1	5
Nickel	4·8	0·7	8·9
Manganese	12·6	0·25	1·8
		0·15	2·1
		0·1	9·2
		0·2	4·0
Chromium	2·9	0·7	9·7
		0·5	5·0
Carbon	0·0045	1	4·95

The gamma rays are scattered frequently in passing through a large bulk of material. However, most of the effect after penetrating a very thick layer is due to photons that have traversed most of the distance from the source in a single long free path, as illustrated in Fig. 2, or with comparatively

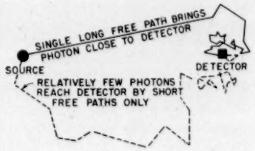
SINGLE LONG FREE PATH BRINGS PHOTON CLOSE TO DETECTOR
SOURCE
DETECTOR
RELATIVELY FEW PHOTONS REACH DETECTOR BY SHORT FREE PATHS ONLY

Fig. 2. Migration of gamma rays through a very thick shield

few small angle deflexions. In spite of the fact that a very long free path is relatively improbable, the photons that have only short free paths between scattering collisions, as illustrated by the dotted line, contribute very little energy to the total penetration, owing to the greater length of their tortuous paths, and the loss of energy in the many collisions.

Mean free path between collisions depends on the energy of the gamma-rays as illustrated in Fig. 3. A single long free path traversing most of the distance from source to detector, as in Fig. 1, most probably occurs at that energy for which the mean free path λ is greatest. The probability of traversing a distance r in a single free path is $e^{-r/\lambda}$. Taking the inverse square law into account the number of gamma-rays reaching a unit area at a distance r from a point source q without scattering is

$$\frac{q}{4\pi r^2}e^{-r/\lambda} \qquad (2)$$

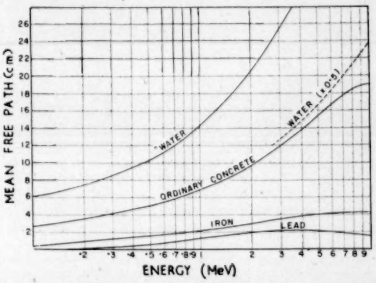

Fig. 3. Variation of mean free path of gamma rays with energy

Including the contribution to the penetration by the photons that are scattered once, twice and more often increases the penetration by a factor B which is called the build-up factor. Hence the total number of gamma-rays reaching a distance r from the source is

$$\frac{q}{4\pi r^2}e^{-r/\lambda}B \qquad (3)$$

The build-up factor B depends on the thickness and kind of material and the initial energy of the gamma-ray quanta. Fairly comprehensive data on build-up factors may be found in papers on radiation shielding.[4]

The build-up factor B usually pertains to the dosage rate (r/h). Hence expression (3) does not represent the actual flux. It gives that flux of photons of the original energy before scattering which would produce the same dosage rate as actually occurs. It is therefore what is required for the simple treatment given here in which the gamma-ray effects are derived from the penetration of the non-scattering primary rays.

In general the source is extended, and it is usual in approximate calculations to treat it as an infinite plane and estimate the intensity at a distance x measured perpendicularly from that plane. The intensity at a distance x from a plane source of strength q per unit area is

$$I = q\int_{x/\lambda}^{\infty}\frac{e^{-y}}{y}dy \cdot B \qquad (4)$$

In practical cases x/λ is usually large and we can make the approximation

$$I = q\int_{x/\lambda}^{\infty}\frac{e^{-y}}{y}dy \cdot B \sim q\frac{e^{-x/\lambda}}{x/\lambda}B \text{ when } \frac{x}{\lambda} \geqslant 1 \qquad (5)$$

Where the radiation penetrates a number of layers of

different shielding materials of thickness t_n and mean free path λ_n the term x/λ should be replaced by

$$\Sigma_n t_n/\lambda_n \tag{6}$$

where the mean free paths λ_n are those characteristic of the gamma-ray energy for which $\Sigma_n t_n/\lambda_n$ has the smallest value, i.e. the energy which gives the greatest penetration.

Fast neutrons are scattered many times before reaching thermal energies and the average angle of scattering is large. Their migration, therefore, can be regarded as a diffusion process in regions close to the source. At greater distances from the source simple diffusion theory does not give a satisfactory approximation in representing their motion. Better approximations are obtained by treating them in the same way as the gamma-rays. Thus the intensity of the neutrons of most penetrating energy at a great distance x from the infinite source is approximately

$$q\frac{e^{-x/\lambda}}{x/\lambda} \tag{7}$$

The source strength q should, of course, include only those neutrons whose energies are in the band of high transmission and above it. The dependence on neutron energy of the mean free path λ in water and in iron is shown in Fig. 4.

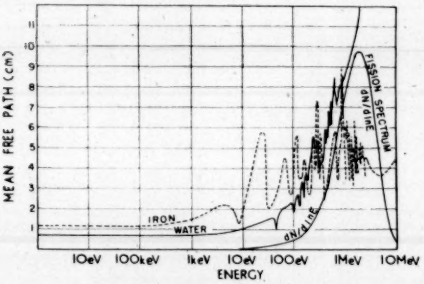

Fig. 4. Variation of mean free path of neutrons with energy

Lower energy neutrons would be rapidly attenuated and would give relatively little contribution to the neutron intensities in the outer part of the shielding. In the case of iron, very fast neutrons have shorter mean free paths than those of energies of a few mega-electron volts. However, these are also available for penetration of the iron at the energies of maximum mean free path when their energies have been reduced by scattering nearer the source.

The fast neutrons, as they lose kinetic energy in collision, produce a thermal neutron flux which is given approximately by

$$\phi = q\frac{e^{-x/\lambda}}{x/\lambda}\frac{\lambda_0}{\lambda}\frac{1}{(1-L^2/\lambda^2)}e^{L_s^2/\lambda^2} \tag{8}$$

where λ is the mean free path of the most penetrating fast neutrons, λ_0 is the capture mean free path of the thermal neutrons, L^2 is the thermal diffusion area and L_s^2 is the migration area for slowing to thermal velocity.

Expression (8) for the thermal flux applies only when the thermal diffusion length L is less than the mean free path λ of the fast neutrons, which is true of most shielding materials. Most of the thermal neutrons in the outer shielding reached the outer layers while they were fast neutrons.

When L is greater than λ, most of the thermal neutrons penetrating the layer of material are ones that entered it as thermal neutrons. This is true in the reflector that surrounds the core of some reactors. In such a case the approximation (8) is not applicable. Diffusion theory should be used.

Expression (8) for the thermal neutron flux is not valid near the boundary between two different materials. Three group diffusion theory or more elaborate methods are required for accurate description of these boundary effects.

Some of the effects that occur near the boundary between iron and concrete shielding are illustrated by some measurements of Bell, Miller and Robson[5] which are plotted in Fig. 5. The thermal neutron flux in the iron is approximately proportional to the fast neutron flux between A and B as predicted by the formula above. The thermal flux in the concrete is also approximately proportional to the fast neutron flux beyond the point D at some distance from the boundary. Transition effects are evident between B and D.

The thermal neutron flux rises between B and C because the neutrons are much more strongly captured in the iron than in the concrete. Thermal neutrons diffuse back from the concrete into the iron contributing to the neutron flux in that material.

The thermal flux between C and D is relatively high because iron is a much poorer moderating material than concrete. Neutrons of intermediate energies migrate from the iron to the concrete before they lose much of their kinetic energy. They are rapidly decelerated in the concrete providing an additional source of thermal neutrons that is important between the boundary and the point D.

It may be seen from a comparison of the curves for gamma-ray intensities and thermal neutron flux that most of the gamma-radiation in the outer parts of the shielding is produced by neutron capture in the region B and C.

The measurements which are plotted in Fig. 5 were made with blocks of iron and concrete placed in one of the experimental holes of the NRX reactor shielding. They were

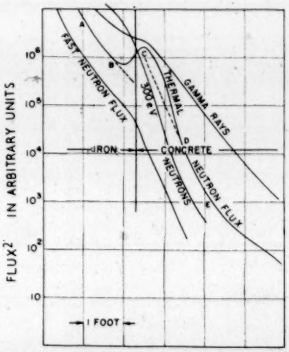

Fig. 5. Neutron flux near boundary between iron and concrete as measured in the NRX reactor shielding

greatly affected by leakage of neutrons and gamma-rays through the cracks around the blocks. The curves of Fig. 5

should not be used therefore for accurate prediction of effects in other shielding.

A convenient approximation in shielding calculations is to regard the reactor as an infinite plane source located at the outer boundary of the core. Thus if $s(t)$ is the source strength (number of photons or neutrons emitted per cm^3 per second) at a depth t below the boundary and k is the absorption coefficient in the core materials, the approximately equivalent source strength at the outer boundary is

$$q = \int_0^\infty s(t)e^{-kt}dt \qquad (9)$$

The contribution to q by radiation originating at depth t decreases rapidly as t increases, and therefore simplifying approximations are easily made in evaluating the integral.

The first step in designing the shielding is to estimate the heating effects produced by the absorption of radiation in structural parts including the shell of the reactor vessel. In water-cooled power reactors, the vessel shell may be of steel several inches in thickness. Absorption of the radiant energy in the steel may result in a much higher temperature in the interior of the wall than at its outer surfaces, causing excessive thermal stresses unless it is prevented by radiation shielding between the core and the shell. A convenient form of shielding is plates of steel immersed in water to cool them. Heavy water may provide the cooling in a heavy-water moderated reactor.

Outside the shell the radiation is still intense enough to produce excessive heating or radiation damage in such common shielding materials as concrete or masonite. The inner layers of the shielding, therefore, are constructed of materials that are resistant to radiation damage and are easily cooled. This part of the shield, which is called the thermal shield, may consist of water, or of steel cooled with water passing through suitable channels, or a combination of both.

In calculating the heating effects it may be assumed for simplicity that each primary gamma-ray quantum and each thermal neutron absorbed contributes an energy of about 8 MeV, or $1{\cdot}4 \times 10^{-11}$ ergs to the shield material. This assumption errs on the side of caution. The effects of the kinetic energy of the fast neutrons are comparatively negligible in the shield because most of their kinetic energy is already lost, and only the energy released by their capture as thermal neutrons need be considered. The energy absorbed per unit volume is therefore

$\phi N\sigma$ ($1{\cdot}4 \times 10^{-11}$ ergs) for thermal neutrons

and I/λ ($1{\cdot}4 \times 10^{-11}$ ergs) for hard gamma-rays.

Using c.g.s. units throughout, these expressions give the heat production in ergs $cm^{-3}\,s^{-1}$. One erg $cm^{-3}\,s^{-1} = 0{\cdot}00965$ B.t.u. $h^{-1}\,ft^{-3}$.

Since the absorption of neutrons results in the production of very penetrating gamma-rays it is desirable to reduce the neutron flux as rapidly as possible in the inner layers of the thermal shield. The choice of a material for this purpose depends on cost, mechanical simplicity, and whether it is more important to save weight or thickness of shielding material. Water (on which useful data has been published[6]) is particularly satisfactory in saving weight owing to the very high scattering cross-section of hydrogen for neutrons of energy below 1 MeV. Where small thickness is desired, water alone does not provide the most effective shield for neutrons because the mean free path in water for neutrons of very high energy, for example energies greater than 5 MV, is longer than in a number of other common materials, including most common metals. For example, see Fig. 4. A combination of water and steel or lead or some other metal provides more rapid attenuation for the whole spectrum of fission neutrons. The most suitable thicknesses for these layers may depend on a number of factors. It may be desirable to provide more metal on the outer regions of the thermal shield than in the inner, because the metal is a much more effective absorber for the gamma-radiation produced by neutron capture in the shielding itself.

Compounds of lithium or boron may be added to the water in the thermal shield to absorb thermal neutrons without producing high energy gamma-radiation. Practical considerations such as increased corrosion, however, may outweigh the advantage.

The part of the shield outside the thermal shield is called the biological shield, because it reduces the intensity of the radiation to a safe level for personnel working near the reactor. It is usually much thicker than the thermal shield. To avoid excessive heating or radiation damage, the rate of energy absorption should not exceed

10^4 ergs $cm^{-3}\,s^{-1}$ in concrete
10^3 ergs $cm^{-3}\,s^{-1}$ in wood, masonite and phenolic products
10^2 ergs $cm^{-3}\,s^{-1}$ in most plastics.

Most of the gamma-radiation entering the biological shield results from neutron capture in the thermal shield and the reactor vessel wall. Usually gamma-radiation produced by fission in the core is relatively unimportant in the biological shielding. In calculating the intensities of these gamma-rays, it may be assumed for simplicity that the capture of a thermal neutron in water produces a gamma-ray quantum of 2·3 MeV, in graphite of 4·5 MeV, and in iron and most other materials of 7·5 MeV.

In the biological shield, since heating effects and neutron damage are unimportant, a wider choice of materials can be considered. The simplest and, in most cases, the lightest biological shield is a large volume of water in which the reactor and thermal shield are immersed. The neutrons are attenuated more rapidly than the gamma-rays in the water and the required thickness is determined by the penetration of gamma-rays from the thermal shield. In contrast, the required thickness of a shield that contains no hydrogen, such as one composed entirely of metal, is determined by the penetration of the neutrons. Saving in weight and thickness is obtained by using a material containing both hydrogen and heavier elements in suitable proportions. Ordinary concrete is very suitable for this purpose and is convenient and inexpensive.

The hydrogen content of most kinds of Portland Cement concrete is greater than 5% of the number of atoms, and this is sufficient to ensure that the biological effectiveness of the neutrons emerging from the outside of the shield is unimportant in comparison with that of the gamma-radiation.

Special kinds of aggregate material may be used in the concrete to increase its effectiveness in absorbing gamma-rays. The rate of attenuation of the gamma-radiation in the concrete is approximately proportional to its specific gravity. Crushed rock of high density or steel punchings or other high density material may be used to increase the density of the concrete, but since the quantities required are large the cost and proximity to the source will be major considerations in deciding to use a special aggregate.

For special purposes, such as mobile reactors, where small

bulk and weight are required, the whole shield may be made of layers of metal and hydrogenous materials. For example, layers of lead or iron might be used with layers of water, masonite or other organic materials. The best relative proportions would have to be determined in each case.

Great care is necessary in the design of reactor shielding to avoid the escape of radiation through cracks, pipe chases and other openings and through large blocks of material embedded in the shielding that are poor shielding material. For example, a one-inch steel bar embedded in the concrete biological shield with its axis in the direction of propagation of the radiation is a channel through which fast neutrons can penetrate much more easily than through the surrounding concrete. If the bar is long the radiation intensity at its outer end may be excessive. Methods for estimating the leakage of radiation through narrow passages are given in a number of published reports, for example reference (2).

The liquid or gas that is used for cooling the reactor becomes radioactive by the capture of neutrons producing radioactive nuclides. The cooling fluid passes through piping, heat exchangers, pumps and other equipment outside the reactor. These need shielding also.

Pumps and other equipment which require maintenance may be placed in rooms with walls designed to give the necessary shielding, if these rooms are kept locked while the plant is operating. However, the radiation may continue to be hazardous during periods of shutdown owing to residual activity in the coolant or the deposition of radioactive impurities on inner surfaces of the equipment. It may be desirable, therefore, to provide local shielding directly on the equipment also. An alternative measure is to limit the time in which personnel are permitted to remain in the room.

Table 2 lists the important radioactive isotopes that are produced in some of the common coolants.

In a closed circuit cooling system the concentration of radioactive nuclides is given approximately by the following expression

$$n \sim N\sigma\phi\tau(1 - e^{-T_e/\tau})\, e^{-T_d/\tau}/(1 - e^{-T_c/\tau}) \qquad (10)$$

where N is concentration of parent atoms
n is concentration of product atoms
ϕ is average neutron flux
σ is activation cross-section
τ is decay period of product atoms
T_e is duration of exposure
T_d is time to flow from reactor to equipment
T_c is time to circulate once.

For a single pass cooling system the concentration of radioactive nuclides is given by

$$n \sim N\sigma\phi\tau(1 - e^{-T_e/\tau})e^{-T_d/\tau} \qquad (11)$$

The number R of radioactive atoms discharged per second in the effluent of a single pass cooling system is

$$R = VN\phi\sigma\tau(1 - e^{-T_e/\tau})e^{-T_d/\tau}/T_e \qquad (12)$$

where V is the volume exposed to the neutron flux.

The presence of impurities can contribute greatly to the radioactivity of cooling liquids. In water or heavy-water closed-circuit cooling systems, the concentration of these impurities can be kept to a very low level by the use of ion exchange resins and filters. The most important radioactive impurities that may occur in a closed water-cooling system are ^{24}Na from aluminium and ^{59}Fe and ^{55}Mn from stainless and other kinds of steel. Where water is used from lakes and rivers for single pass cooling the dissolved minerals in the water can produce a very high radioactivity in the effluent.

In some reactors there are spaces in regions of high neutron flux that contain air. Argon-41 is produced and may be an important health hazard if it is allowed to leak into the operating room. This can be prevented by drawing the air through suitable shielded ducts and discharging it from a high stack. Under some meteorological conditions, such as the existence of temperature inversions, the radioactive air discharged from the stack may remain stagnant over the plant site. The time of exposure T_e is usually short compared with the decay period (1·8 h/0·69 = 2·6 h) of ^{41}A and therefore

$$R \sim VN\phi\sigma \qquad (13)$$

Hence the volume V of the air spaces in the reactor should be kept small. R is almost independent of the rate of flow V/T_e of air through the spaces.

The design of the shielding for nuclear reactors is a complex, laborious and detailed undertaking. There are many uncertainties in the data. Approximation must be made in the calculations if they are to be completed in reasonable time. It is easy to overlook important effects. To allow for error, it is prudent when the design is completed to increase the

Table 2. *Radioactive isotopes produced by coolants*

Coolant	*Nuclear reaction*	*Cross-section*	*Half-life*	*Threshold*	*Quanta per capture*	*Radiation emitted*
Air	^{40}A$(n\gamma)^{41}$A	0·626	1·8 h		1 1	1·25 MeV β 1·3 MeV γ
Water including D_2O	^{16}O$(np)^{16}$N	0·027	7·5 s	11 MeV*		7·1 MeV γ 10 MeV β
	^{17}O$(np)^{17}$N	0·020	4·14 s	11·2 MeV*		1 MeV n
	^{18}O$(n\gamma)^{19}$O	0·000 22	29·4 s		0·7	4·5 MeV β 1·6 MeV γ
	^{2}H$(n\gamma)^{3}$H	0·0003	12·5 yr			
Sodium	^{23}Na$(n\gamma)^{24}$Na		14·8 h			β
Water	^{27}Al$(n\alpha)^{24}$Na		14·8 h			no γ-rays
Water	^{27}Al$(n\gamma)^{28}$Al	0·21	2·4 min			
Water	^{58}Fe$(n\gamma)^{59}$Fe	0·36	46 days		0·5 0·5	1·1 MeV γ 1·3 MeV γ
Water	^{54}Mn$(n\gamma)^{55}$Mn	13				

* The fraction of the fission neutron spectrum above these thresholds is $2{\cdot}2 \times 10^{-4}$.

thicknesses of reactor materials. The surest and most satisfactory approach is to base the design as far as possible on experience of the shielding of other reactors already in operation or on tests made with a mock-up of the proposed reactor shield. Even if this is done the designer is faced with many problems in estimating the possible leakage of radiation through pipe-chases, apertures around embedded equipment and experimental facilities, control devices and the channels through which the reactor fuel is replaced.

ACKNOWLEDGEMENT

Reports of shielding calculations by W. J. Henderson, A. C. Whittier and I. L. Wilson, which have not yet been declassified because they also contain secret information, have been of great assistance to the author in preparing this paper.

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The control of reactors

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The concepts of reactivity, neutron lifetime and delayed neutrons are described and their relationships with the rate of change of neutron density and reactor power are discussed. The amounts of reactivity to be controlled in a reactor may be large and varying with time. The various components of this reactivity are noted and discussed. The physical methods of control are surveyed and the safety of reactors is briefly considered.

INTRODUCTION

A nuclear reactor may be part of a power system, an experimental prototype of a power reactor or a research reactor, perhaps of very low power output, acting simply as a radiation source. In each case the reactor has to be controlled so that, at least, it can be set at some required power level, maintained at that power and then shut down. Also it is usually convenient to be able to bring the reactor up to power any time after it has been shut down. The possibility of the reactor getting out of control must be minimized so far as is reasonable and, further, the possibility of damage to the reactor must be reduced to a minimum if it does get out of control. This paper briefly discusses the physics of reactors so far as it is pertinent to this general problem of the control of reactors.

Throughout the paper reference is made to reactors of several types which are denoted by generally accepted abbreviations. References are normally given for data concerning these reactors. However, in the case of the gas-cooled graphite-modulated reactors which will be the initial type used for power production in the United Kingdom, and denoted in the paper by the name PIPPA, any data quoted is only an estimate based on information generally available. This data will, nevertheless, give an indication of the behaviour of this reactor type.

FUNDAMENTAL CONCEPTS

A list of symbols will be found in the appendix

The thermal power produced in a reactor as a prompt result of the fission process is related to the neutron density by the expression:

$$P_p = \int_V \int_v n(v,V) \sum_f (v) p dv dV \qquad (1)$$

where p is the prompt power equivalent to one fission per second. It is not possible to give simply an exact expression relating this thermal power to the neutron density. This is because the macroscopic fission cross-section is dependent on the reactor design; for example, it will depend on the enrichment of the fuel and the ratio of fuel to moderator. Further, the neutron density will have a continuous velocity spectrum which again depends on the reactor design, the mean neutron velocity increasing in magnitude as we consider the range of reactors from the thermal type through the intermediate to the fast reactor. Even within the thermal group of reactors the mean of neutron velocity will depend on the operating temperature and it will in fact vary within one reactor as it starts operation from the cold state and rises to its normal designed temperature.

Let us assume that the neutron density maintains its space distribution constant as a function of time in the reactor so that it can be specified by a mean neutron density and a form factor; also that the reactor is operating under such conditions that the neutron density, $n(v)$ can be characterized by a velocity v. Then we have:

$$P_p = nv.p.\sum_f (v) \int_V a(V) dV \qquad (2)$$

nv, is, of course, a representative neutron flux which can perhaps best be regarded as the total neutron track length per cubic centimetre per second. $\sum_f (v)$ is the probability per centimetre that a neutron of velocity v will produce a fission. Thus from expression (2) we see that the reactor power immediately due to the fission process is directly proportional to the neutron density.

The energy released by one fission is distributed[1] as shown in Table 1. The neutrinos do not react with matter to any appreciable extent and so essentially escape from the reactor without releasing within it their energy. Thus, to the

accuracy of the data available, 200 MeV per fission are released within the reactor. $3{\cdot}1 \times 10^{10}$ fissions per second thus provide throughout the reactor a thermal power of 1 W.

Table 1. *Distribution of energy from thermal fission of* ^{235}U

Kinetic energy of fission fragments	169 MeV
Kinetic energy of fission neutrons	5
γ-radiation directly from fission	5
γ-radiation from fission products	7
β-radiation from fission products	5
β- and γ-radiation from neutron captures	8
Neutrinos	9
Total, prompt and delayed	208 MeV

The 200 MeV per fission is not all released instantaneously, the β and γ-radiations from the fission products will be released some time later. These fission products constitute an intense source of radioactivity and appreciable thermal power if the reactor has been operating for some time at power. At the initial start-up of the reactor none of this energy will be available and we shall observe only about 187 MeV per fission. However, after operation at power for some time the radioactivity will approach its equilibrium value and the energy release per fission will approximate to 200 MeV. After shut-down of the reactor, as we shall discuss later, power will continue to be released as the accumulated fission products and any other accumulated radioactive isotopes decay.

Reactivity. A reactor normally operates at a steady power level when the number of neutrons produced by the fission process in one generation within the reactor is equal to the corresponding number in the previous generation. The ratio of these two numbers is termed the effective multiplication factor for the reactor. In this state the reactor is said to be critical.

If k_{eff} is less than unity the reactor is said to be sub-critical and the power will normally decrease. In practice, k_{eff} may range from about 0·96 to about 1·001 during normal operation, in some cases it may be less than 0·9 when the reactor is fully sub-critical. If k_{eff} is greater than unity the reactor is said to be super-critical and the power will increase. $(k_{eff} - 1)$ is termed the excess multiplication factor. The reactivity of a reactor is defined as:

$$\rho = (k_{eff} - 1)/k_{eff} \simeq k_{ex} \tag{3}$$

as k_{eff} is near unity.

The multiplication factor which would occur if the core of the reactor extended to infinity is denoted by k_∞. This may range from about 1·03 to almost 2, depending on the state of the reactor and its design. The maximum possible value of k_{eff} will be less than k_∞ by the fractional number of neutrons per generation lost by leakage from the finite reactor. The difference between the desired and the maximum possible value of k_{eff} will, in general, have to be accommodated by a control system.

REACTOR KINETICS

Let us consider the simplest possible case of changing neutron density. This occurs when all the neutrons per fission are considered to be emitted at the moment of fission. Then:

$$dn/dt = (nk_{eff} - n)/\tau \tag{4}$$

Thus
$$n(t) = n(o) \exp\left[(k_{eff} - 1)t/\tau\right] \tag{5}$$

and the reactor period, normally defined as $n/(dn/dt)$, which in this case is the time taken to increase the neutron density by a factor $e = 2{\cdot}716$, is:

$$T = \tau/(k_{eff} - 1) \tag{6}$$

In this simple case the reactor power thus continuously increases or decreases exponentially with time, the period depending on the neutron lifetime and the excess multiplication factor.

The neutron lifetime τ is the sum of the average times spent in slowing down and in thermal diffusion. In general, the slowing down time in a thermal reactor is small compared with the diffusion time; in a fast reactor there is, of course, no thermal diffusion time. Examples of approximate neutron lifetimes in various reactors, with their components, are given in Table 2.

It will be seen that with a reactivity of about 0·1%, i.e. 10^{-3}, the periods of all the reactors noted in Table 2 are of the order of one second or smaller. From the control viewpoint this is an exceedingly small time. However the natural phenomena of delayed neutrons come to our assistance to make the reactor periods effectively longer.

Delayed neutrons and their effects. The neutrons propagating the chain reaction may come from a nuclear fission, an artificial or spontaneous neutron source in or near the reactor or from radioactive fission products. The neutrons from fission are emitted a very short time, about 10^{-14} s, after the primary neutron has been absorbed in the fissile nucleus; such neutrons are termed "prompt" neutrons and correspond to the type we have discussed in the previous section. The neutrons from the fission products are emitted some time after the fission event, they are therefore termed "delayed" neutrons. The actual time interval between fission and the emission of a delayed neutron will depend on the neutron-emitting fission product and its position in a decay chain. Six delayed neutron groups have been definitely observed in the fission products. There is some evidence that there are further groups of very low relative intensity. One would expect that the decay constant of a group λ_i will be the same whatever the fission event. This appears to be confirmed by experiments. However, the yield neutrons from each group expressed as a fraction of the total fission yield, β_i, will depend on the fission event, differing with the nuclei fissioned. To a lesser event it will depend on the

Table 2. *Neutron lifetimes in representative reactors*

Reactor	*Moderator*	*Fuel*	l_s		τ
PIPPA	Graphite	Nat. U	10^{-4} s	9×10^{-4} s	10^{-3} s
DIDO	Heavy water	Enriched U	5×10^{-5}	10^{-3}	10^{-3}
MERLIN	Water	U, 20% ^{235}U	10^{-5}	$5{\cdot}3 \times 10^{-5}$	$6{\cdot}3 \times 10^{-5}$
ZEPHYR	—	^{239}Pu	$\sim 10^{-8}$	0	$\sim 10^{-8}$

energy of the neutron producing the fission. The half-life-times of the groups and their decay constants, as observed for thermal neutron induced fission in ^{235}U, are given in Table 3.[2] It will be noted that the various delayed neutron groups and the prompt neutrons all have different energies, thus their effect in a reactor will be slightly different.

Table 3. *Delayed neutron groups*

Group,	*En, keV*	*Half-life, s*	λ_i, s^{-1}
1	250	55·6	0·0125
2	570	22·0	0·0315
3	412	4·51	0·154
4	670	1·52	0·456
5	400	0·43	1·61
6	—	0·05	13·9

Other groups with either very short or rather long half-lives and all of low relative intensity have been reported; however, from the control viewpoint they are unimportant. The half-lives and the fractional yields of the delayed neutron groups from the fission of the various isotopes, Table 4, are still open to some doubt. The values listed in Tables 3 and 4 give reasonable practical agreement with observed reactor behaviour. A comprehensive and up-to-date account of delayed neutron measurements has been given by Keepin.[3] From Table 4 it will be noted that in a thermal spectrum ^{235}U has a total delayed neutron fraction at least double that of either ^{233}U or ^{239}Pu. In a reactor moderated by heavy water there are further sources of delayed neutrons. These arise from photo-disintegrations of the deuterium by the γ-radiation from the fission products. The decay constants and yields of nine delayed neutron groups arising in this way have been measured.[4]

Table 4. *Fractional yields of the delayed neutron groups*

	$\beta_i \times 10^3$						
	Thermal			*Fast*			
Spectrum Isotope	^{233}U	^{235}U	^{239}Pu	^{232}Th	^{235}U	^{238}U	^{239}Pu
Group 1	1·8	2·5	1·4	8	2·5	2	0·85
2	5·8	16·6	10·5	33	14	22·5	6
3	8·6	21·3	12·6	41	13	32	4·5
4	6·2	24·1	11·9	114	27·5	70	8·5
5	1·8	8·5		42	9	41·5	2·5
6	—	2·5	—	8·5	1	6	0·15
$\beta \times 10^4$	24·2	75·5	36·4	246·5	67	174	22·5
ν assumed				2·55	2·57	2·52	3·01

Let us now consider the effect of the delayed neutrons on the reactor behaviour. The neutron density equation, corresponding to expression (4) for prompt neutrons only, now becomes:

$$dn/dt = k_{eff}.n/\tau - \beta n/\tau + \sum_{i=1}^{m} \lambda_i C_i \qquad (7)$$

where

$$dC_i/dt = \beta_i n/\tau - \lambda_i C_i \qquad (8)$$

Here the total rate of change of neutron density is equal to the prompt rate less the delayed neutron fraction, also the rate of build-up due to the delayed neutrons from past fissions has to be added. The equations represented by equations (7) and (8) can be solved for various reactivity input functions. The solution for a step reactivity input is characterized by an equation of the form:

$$\rho = \tau\omega/k_{eff} + \sum_{i=1}^{m} \omega\beta_i/(\omega + \lambda_i) \qquad (9)$$

This relates the step reactivity input ρ with the reactor design and the $(m + 1)$ values of ω which it denotes. The neutron density is then given by:

$$n = n_0 \sum_{=0}^{m} A_j e^{\omega_j t} \qquad (10)$$

where ω_0 has the same sign as ρ and the other m exponents are negative. The inverse times, excepting the first, are given approximately by:

$$\omega_j = -\lambda_{i=j-1} \qquad (11)$$

The constants A_j are determined by the initial conditions in the reactor. If the reactivity step is positive the negative exponents eventually lead to a decrease in the importance of their terms and soon the neutron density is set essentially by the exponent ω_0. This occurs after a time the order of $1/\lambda_1$, then:

$$n = n_0 A_0 e^{\omega_0 t} \qquad (12)$$

We see that the reactor period, T, is then $1/\omega_0$; this is termed the stable period.

To investigate the effects of various reactivity inputs on the neutron density either a digital or an analogue computor is normally used. If neither of these instruments is available approximations to the effect of the delayed neutrons can be made to obtain the order of the effects. Glasstone gives several examples of a method which assumes the delayed neutrons are a single group with a decay constant which is the mean of the decay constants of the actual delayed neutron groups.[5] Schultz[6] gives examples of a treatment, for step, ramp and sinusoidal reactivity inputs, in which the delayed neutrons are treated as a constant source for the first 0·1 seconds after the initiation of the disturbance. This gives a good approximation to the initial portion of the neutron density response. The effects of step reactivity inputs are shown in Fig. 1 for a PIPPA type reactor and a small light water moderated reactor.

Writing expression (9) with $1/\omega$ replaced by T, we have the reactivity related to the stable reactor period:

$$\rho = \tau/(Tk_{eff}) + \sum_{i=1}^{m} \beta_i/(1 + \lambda_i T) \qquad (13)$$

The stable periods of the reactors in Fig. 1, as well as for a fast reactor, all using ^{235}U as fuel, are shown in Fig. 2 for various positive step changes in the reactivity. It can be seen that for small values of reactivity the stable period is insensitive to the neutron lifetime. Further, the periods are

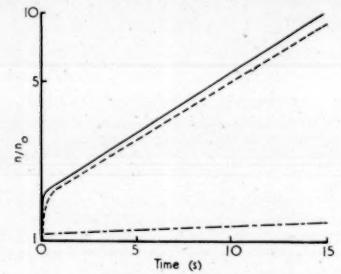

Fig. 1. Relative neutron density as a function of time for various step reactivity inputs. Fuel ^{235}U

——— $\rho = 0{\cdot}001$, $\tau = 10^{-3}$ s.
——— $\rho = 0{\cdot}003$, $\tau = 6{\cdot}3 \times 10^{-5}$ s.
– – – – – – $\rho = 0{\cdot}003$, $\tau = 10^{-3}$ s.

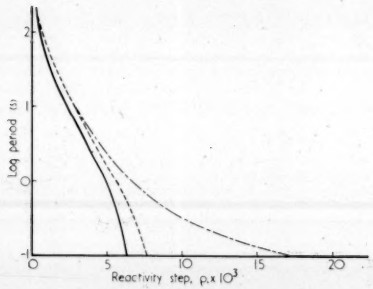

Fig. 2. The stable periods of reactors with various lifetimes for positive step reactivity inputs

——— $\tau = 10^{-7}$ s.
– – – – – – $\tau = 6{\cdot}3 \times 10^{-5}$ s.
— — — — — $\tau = 10^{-3}$ s.

now much longer than they were without the delayed neutrons thus making the control of reactors possible with reasonable effort. The effect of the delayed neutrons is shown in Table 5. As would be expected the smaller fraction of delayed neutrons

Table 5. *The stable period of reactors with and without delayed neutrons. Step reactivity input* $= 10^{-3}$

τ	10^{-3} s	$6{\cdot}3 \times 10^{-5}$ s	10^{-7} s
	s	s	s
No delayed neutrons	1	$6{\cdot}3 \times 10^{-2}$	10^{-4}
With delayed neutrons from ^{235}U	90	90	45
With delayed neutrons from ^{239}Pu	25	25	7·4

from ^{239}Pu results in a shorter stable period for a reactor fueled with ^{239}Pu compared to a reactor fueled with ^{235}U.

For large reactivities the stable period is given by:

$$\rho = \tau/(Tk_{eff}) + \sum_{i=1}^{m} \beta_i, \qquad \lambda_i T \ll 1 \quad (14)$$

Thus we have:

$$T = \tau/[k_{eff}(\rho - \beta)]$$
$$= \tau/[(\rho - \beta)(1 + \rho)], \text{ approximately} \quad (15)$$

When $\rho \gg \beta$ we have simply:

$$T = \tau/(k_{eff} - 1)$$

which is the same as if all the fission neutrons were prompt. Thus for large reactivity steps the period decreases in proportion to the neutron lifetime. When the reactivity is equal to the delayed neutron fraction the reactor is said to be prompt-critical, when the reactor is above prompt-critical expression (15) can be used to obtain the stable period, Fig. 3.

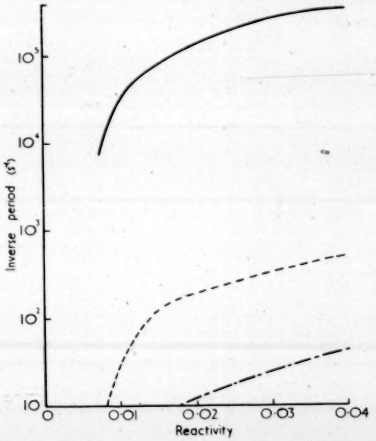

Fig. 3. Kinetic behaviour above prompt critical. Inverse period as a function of reactivity. Fuel ^{235}U

——— $\tau = 10^{-7}$ s.
– – – – – – $\tau = 6{\cdot}3 \times 10^{-5}$ s.
— — — — — $\tau = 10^{-3}$ s.

Summarizing, at small reactivities all reactors behave rather similarly from the control viewpoint. As the reactors become prompt-critical their behaviour becomes very different, those having a short neutron lifetime have a correspondingly rapid response leading to great rates of power rise. This is especially true of the fast reactors. One of the aims of reactor design must be to minimize the possibility of the reactor inadvertently getting near or beyond prompt-critical.

Neutron sources at start-up. Assume that a source of neutrons is placed in the reactor core. The kinetic equations are now:

$$dn/dt = (k_{ex} - \beta)n/\tau + \sum_{i=1}^{m} \lambda_i C_i + S \quad (16)$$

$$dC_i/dt = \beta_i n/\tau - \lambda_i C_i \quad (17)$$

If the reactor is sub-critical, so that k_{ex} is negative, and has taken up some steady state so that:

$$dn/dt = 0 \text{ and } dC_i/dt = 0 \tag{18}$$

then it follows that:

$$n = -S\tau/k_{ex} \tag{19}$$

The reactor is now acting as a neutron multiplier, the multiplication factor being $(1 - k_{eff})^{-1}$. For example, when k_{eff} is 0·95 this multiplication factor is 20.

One of the basic principles of reactor control is that if the reactor is in such a state that it could reasonably become critical then it should be possible to observe the prompt reactor power by means of the reactor instrumentation. This will be especially necessary when the reactor is being taken to the critical state, i.e. when reactivity is being added. The behaviour of the reactor described by expression (19) helps to make this a reasonable task. Wherever there is fissile material there is a neutron source due to spontaneous fission neutrons; e.g. the spontaneous fission power of one gramme of natural uranium is $2{\cdot}2 \times 10^{-13}$ W. A large gas-cooled graphite moderated reactor may contain as much as 200 tons of natural uranium and hence there is a built-in neutron source with a thermal power of about 5×10^{-5} W. If the shut down reactivity of this reactor is -2×10^{-2} then the steady shut-down power, due to neutron multiplication, is about $2{\cdot}5 \times 10^{-3}$ W. If the peak power of this reactor is 500 MW then the range of the instrumentation required for start-up is just over eleven decades. In an enriched fuel reactor the critical mass will be smaller than that for the previous reactor. The amount of uranium in a medium-sized water cooled and moderated research reactor may be about 20 kg if the uranium is enriched to the extent of being 20% ^{235}U. The spontaneous neutron power at start-up would then be only 2×10^{-7} W and if the maximum power of the reactor is 5 MW then the range of control and instrumentation must be over about thirteen decades. This problem can be reduced in magnitude by inserting a neutron source into the reactor core. The source is multiplied by the reactor core and during shut-down an appreciable power is developed which makes the range of control more reasonable. Thus in the research reactor mentioned above the shut-down power, with k_{eff} about $-0{\cdot}05$, will be on average about 3 mW with a consequent range of control over about nine decades.

Table 6. *Neutron sources used to facilitate continuous observation of the neutron density during start-up*

Source	*Half-life*	*Neutron emission/curie/s*	*Associated γ-radiation. (r/h at 1 metre from 1 curie)*
Ra–Be	1620 y	$1{\cdot}7 \times 10^{7}$*	0·8
Po–Be	138 d	3×10^{6}*	negligible
Sb–Be	60 d	$1{\cdot}9 \times 10^{5}$†	1
Na–Be	14·8 h	$1{\cdot}3 \times 10^{5}$†	1·9

* Intimate mixture of α-ray emitter and beryllium.
† 1 g beryllium at 1 cm from γ-ray emitter.

Some of the common neutron sources are given in Table 6. The neutrons are usually produced by an (α, n) or (γ, n) reaction, the initial radiation being emitted by a radioactive isotope mixed with the neutron producing material. Most of these sources have a relatively short half-life, which can be a disadvantage in a zero-energy reactor, and they emit γ-radiation as well as neutrons. This latter fact may make the positioning and/or changing of the sources difficult. An alpha-ray emitter with a long half-life is ^{239}Pu, 24 000 years; however, this is at present difficult to obtain.

If the reactor has been operating for some time then the fission product γ-radiation will produce neutrons by photo-disintegration if there is any deuterium or beryllium in the core region. If such a reactor is operated for long periods at power then it may only be necessary to introduce an artificial source into the core at the initial start-up and when the core is changed.

Reactor shut-down and fission product power. If negative reactivity is applied to a critical reactor then k_{eff} will become less than unity and the neutron density will decrease. All $(m + 1)$ values of ω_j in expression (10) become negative and thus all the terms decrease with time. Eventually the term with the smallest numerical value of ω, ω_0, becomes dominant and the density decreases with the time constant $T = 1/\omega_0$, that of the most delayed neutron group. The form of the neutron density response for various negative step reactivity inputs is shown in Fig. 4. This illustrates the

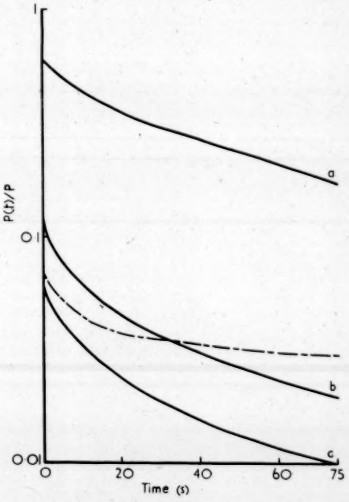

Fig. 4. Relative reactor power due to a negative step reactivity input

Curve (*a*), $\rho = -0{\cdot}005$: Curve (*b*), $\rho = -0{\cdot}055$: Curve (*c*), $\rho = -0{\cdot}11$.

— —— — —— — Fission product power.

constant period of 80 s which occurs after the transients have become small. Unfortunately some of the reactor power arises from the radioactivity of the fission products, thus the reactor power does not decrease to the same extent as does the neutron density. This fission product power decreases with time according to the quantities and decay constants of the various radioactive isotopes produced. It is, in fact, an appreciable portion of the total reactor power and decays relatively slowly with time. If a reactor has been operating at a power P for a time T_0 it has been found that the fission product power, P_D, will be given by:

$$P_D = 0{\cdot}07\, P[t^{-0{\cdot}2} - (t + T_0)^{-0{\cdot}2}] \tag{20}$$

This expression can be used to determine the fission product power for times t greater than one second after the reactor has been fully shut-down. Thus after shut-down the actual reactor power output will be obtained by a combination of the delayed fission power given by expression (20) and the prompt fission power obtained by using an expression similar to (10). Table 7 gives these two effects as a function of time

Table 7. *Reactor power output after shut-down with* $\rho = -0{\cdot}055$

Time after shut-down	*Power output*			
t	$P = 1$ *MW* $\tau = 6.3 \times 10^{-5}$ *s*		$P = 500$ *MW* $\tau = 10^{-3}$ *s*	
	P_p	P_D	P_p	P_D
10 *s*	60 kW	44 kW	30 MW	22 MW
10^2	14	28	7·3	14
10^3	$1{\cdot}9 \times 10^{-3}$	18	$9{\cdot}4 \times 10^{-4}$	8·8
10^4	$<10^{-6}$	11	$<10^{-9}$	5·5
10^5	—	7	—	3·5

for a research reactor and a power reactor which have both been operating at a constant power and then are shut down by the insertion of a reactivity step of $-0{\cdot}055$. The power of the reactor in this case is soon mainly determined by the delayed fission product heat output. It will also be observed that in large reactors this delayed power output can be very appreciable.

METHODS OF CONTROL

If a constant power level is required, then, as stated above, the neutron balance between production and consumption in the reactor must be adjusted so that the number of neutrons propagating the chain reaction in any generation is just equal to that in the preceding generation. This approach neglects the delayed heat release due to fission product radioactivity as usually the delayed power will be smaller than the prompt power and will also usually be associated with a longer period. In a critical reactor then the neutron births and fates will have taken up some steady average state in which for each fission one more fission will be produced, whilst the remainder of the neutrons from the initial fission will suffer a variety of fates not immediately productive of further neutrons. These fates, in their various proportions, shown in Table 8, may be absorption in fertile material, in coolant, in moderator, in other materials in the core such as fuel sheaths, in absorbers with a variable magnitude deliberately introduced, or the neutrons may leak out from the core and be lost to the reaction.

Table 8. *Neutron balances in one fission cycle*

Component	*Na-cooled, graphite moderated power reactor(s)*	*Water cooled and moderated research reactor*
Production	100	100
Absorption in ^{235}U	46·88	48·08
Resonance capture in ^{238}U	22·91	3·28
Thermal capture in ^{238}U	10·87	0·77
Epithermal leakage	5·95	24·35
Thermal leakage	1·41	3·10
Absorption in coolant and moderator	4·60	11·41
Absorption in sheaths and structural material	3·12	3·91
Poisons	2·40	3·1
Control absorbers	1·86	0·5
Experiments	—	1·5
Total consumption	100	100

In order that the reactor may become super-critical the steady neutron balance must be disturbed so that the neutron consumption in either non-neutron productive absorption or leakage is reduced, or the productive absorption is increased. Similarly, to make the reactor sub-critical either the leakage or non-neutron productive absorption is increased or the fissile absorption is decreased. Thus the effective reproduction factor may be varied, and so the reactor controlled, by varying the leakage, the non-neutron productive absorption, or the fissile absorption. Each method has some effect on the other methods, thus if absorbers are inserted into the core then the leakage is changed to some extent, as well as the fissile absorption. This might be expected as it would be rather surprising if the new neutron balance was such that the change in the number of neutrons that took part in further fissions were exactly balanced by the change in the

Table 9. *Methods of control used in reactors*

Reactor type	*Thermal*		*Fast*
Method	*Heterogeneous*	*Homogeneous*	
Productive absorption			
Movement of fuel	MTR	ZETR	ZEUS, Dounreay
Concentration of fuel		HRE	
Non-productive absorption	PIPPA MTR DIDO MERLIN DIMPLE	HRE ZETR	
Leakage			
Movement of moderator	NRX, DIMPLE		
Movement of reflector			ZEPHYR, EBR
Depth of liquid reflector	DIMPLE	HRE ZETR	
Temperature	CP5	HRE	
Voids	EBWR		

non-productive absorption. As a general rule it may be stated that the main method of control used in thermal reactors to date is variation of the non-neutron productive absorption; in fast reactors the main method used is variation of the leakage. The various methods used to achieve control are shown in Table 9 for a selection of reactors. In several reactors more than one variation of a primary method is used; thus in the heavy water moderated and reflected reactor DIMPLE, the variation in depth of the top reflector is used as a fine control whilst the movement of both top reflector and part of the moderator can be used when a large reactivity change is required. In other reactors more than one primary method is used; for example, in the Materials Testing Reactor some of the control units consist of a fuel box attached to a cadmium absorber, thus one unit can vary directly both the productive and non-productive absorption.

Any leakage or absorption in a non-fissile or non-fertile material is a waste of a neutron which could possibly have been used to produce a fissile nucleus. As every effort is usually made in the design of a power reactor to obtain a high conversion factor there will be a tendency to minimize the use of such methods of control. A compromise will have to be made between the economic design of the control system and the economic requirement of a high conversion factor.

The control units. The units in the reactor which are used to control the reactivity are usually discussed in three groups. The fine or regulating units provide a small variation of reactivity such as would be used to maintain the reactor critical when it is operating under relatively stable conditions. The amount of reactivity invested in the fine units is normally between $\beta/3$ and $2\beta/3$. Maximum rates of change of reactivity due to the fine units are usually between 10^{-3} per second and 10^{-4} per second. Coarse control of reactivity is accomplished by means of the coarse, or shim, units. There are usually several of these and they may control, together, as much as 0·2 in reactivity. They are used to take the reactor up to its operating power, shut it down and to compensate for large-scale reactivity changes which occur during operation. A typical rate of reactivity addition by these units is between 10^{-4} per second and 10^{-6} per second.

There are then the shut-down, or safety, units which provide a fast reduction in reactivity and are used to shut-down the reactor rapidly. These are sometimes combined with the coarse units, or with the coarse plus fine units, to form a combined shut-down system. The reactivity controlled by the shut-down system may be considerably greater than the total possible reactivity of the reactor, a factor of up to two being used. All this reactivity is usually capable of being inserted, in a negative sense, within one second or less. If the coarse or fine units are included in the shut-down system they must be capable of either reducing reactivity rapidly or relatively slowly as in normal shut-down and operation.

REACTIVITY CHANGES DURING OPERATION

Fission product poisoning. During operation of the reactor the fission products usually accumulate, to some extent these are all parasitic neutron absorbers and thus are termed "poisons" within the reactor. In a fast reactor this poisoning is not particularly serious. However, two of the fission product isotopes, ^{135}Xe and ^{149}Sm, are produced with a high yield and have very large absorption cross-sections in the thermal neutron energy range.

^{135}Xe occurs mainly in the decay chain arising from the fission product ^{135}Te, $\gamma = 5{\cdot}6 \times 10^{-2}$:

$$^{135}\text{Te}(\tau_{\frac{1}{2}} = 2\text{ min}) \rightarrow {}^{135}\text{I}\ (6{\cdot}7\text{ h}) \rightarrow {}^{135}\text{Xe}\ (9{\cdot}2\text{ h}) \rightarrow$$
$$\rightarrow {}^{135}\text{Cs}(2 \times 10^{4}\text{ yr}) \rightarrow {}^{135}\text{Ba (stable)}$$

The isotope is also produced to a small extent as a direct fission product, $\gamma = 3 \times 10^{-3}$. During operation of the reactor the ^{135}Xe poison is formed by decay of the ^{135}I and is itself lost by radioactive decay and by neutron capture. After the reactor is made sub-critical the loss of ^{135}Xe due to neutron capture will not occur. This results in a different behaviour of the poisoning effect between the critical and sub-critical states. The equations symbolizing these effects are given adequately elsewhere.[8] The resultant reactivity loss, as a function of time, during operation is shown in Fig. 5

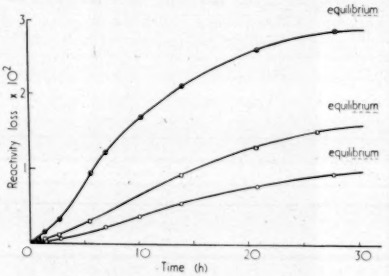

Fig. 5. Effect of xenon poisoning on reactivity as a function of operating time of reactor

○ $\bar{\phi}$th $= 2{\cdot}6 \times 10^{12}$ n cm^{-2} s^{-1}.
□ $\bar{\phi}$th $= 5{\cdot}3 \times 10^{12}$ n cm^{-2} s^{-1}.
⊗ $\bar{\phi}$th $= 2{\cdot}6 \times 10^{13}$ n cm^{-2} s^{-1}.

for a typical thermal reactor operating at various constant mean fluxes. The reactivity loss rises to an equilibrium value which is a function of the thermal flux; these equilibrium values are shown in Fig. 6. For a given flux the reactivity loss due to poisoning is a function of the thermal utilization factor f hence the data presented in the figures can be related to any reactor. After one day of constant operation the

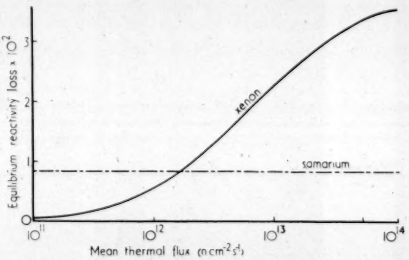

Fig. 6. Equilibrium poisoning effect on reactivity of a reactor ($f = 0{\cdot}75$)

——— xenon — — — — — samarium

xenon poisoning effect is almost at its equilibrium value. After the reactor has been made sub-critical the reactivity loss rises to a maximum in about ten hours; this is shown in Fig. 7. This loss can be very large, amounting to about 0·4 in a reactor such as the Materials Testing Reactor which has a mean thermal flux of 2×10^{14} n cm^{-2} s^{-1}. Normally

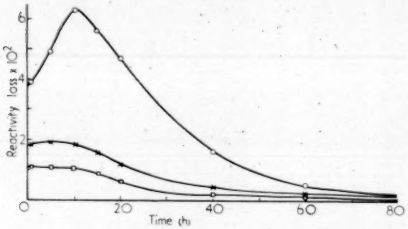

Fig. 7. Reactivity loss due to xenon build-up after shut down. ($f = 0{\cdot}75$)

○ $\bar{\phi}$th $= 2{\cdot}6 \times 10^{13}$ n cm^{-2} s^{-1}.
× $\bar{\phi}$th $= 5{\cdot}3 \times 10^{12}$ n cm^{-2} s^{-1}.
□ $\bar{\phi}$th $= 2{\cdot}6 \times 10^{12}$ n cm^{-2} s^{-1}.

sufficient reactivity would not be available in the core to compensate such large losses, so with a high thermal flux reactor it is desirable that the possibility of inadvertent shut-down be reduced to a minimum and that a fault, if it occurs, can be remedied quickly. However, these requirements, or that of a fast start-up after being sub-critical, must not jeopardize the safe operation of the reactor. Another method of overcoming xenon poisoning in high flux reactors is to remove the gaseous fission products continuously; this will be possible in reactors using a liquid fuel. At the same time the conversion factor will be increased.

The effects of the poisoning due to ^{149}Sm are different because the samarium isotope is stable and so is not lost by radioactive decay; further, the periods involved are much longer.

$$^{149}\text{Nd}(1{\cdot}7\text{ hr}) \rightarrow {}^{149}\text{Pm}(47\text{ hr}) \rightarrow {}^{149}\text{Sm}(\text{stable})$$

The general effect is that the equilibrium reactivity loss, 0·012 maximum, is independent of the mean thermal flux and the time taken to reach this equilibrium value is greater, the order of 100 days in a medium flux reactor, see Fig. 8. After shut-down the peak reactivity loss is reached after about 10 days, Fig. 9.

The majority of the fission products do not have these high capture cross-sections, they can be taken into account by calculating their build-up with time of operation and allotting to them a mean absorption cross-section of about 40 barns per atom in thermal systems.[9] It is estimated that in intermediate systems the loss of reactivity due to the majority of the fission products will be greater than in the thermal systems. Their mean absorption cross-section has been calculated to be about 8 barns per atom at a neutron energy of 100 eV,[10] the corresponding fission cross-section for ^{235}U being 23 barns compared with about 600 barns at thermal energies.

Fuel depletion and conversion. When the reactor is operating fissile material is being "burnt" with the resultant production of fission products. Some of the fissile material is also absorbing neutrons without fission occurring and generally fertile material will be absorbing neutrons to produce, eventually, further fissile material. The chain of events is exceedingly complicated as materials are constantly produced and destroyed in the reactor core. The reactivity available at any time will thus be a complicated function of the neutron fates in the reactor. To a first approximation the reactivity due to a nucleus will depend on $(\nu - 1)\sigma_f - \sigma_c$. Thus the overall reactivity will depend on the cross-sections for the various processes, the values of ν of the fissile materials for the neutron spectrum in the reactor, the flux level and the design of the reactor which sets the value of the conversion factor and its variation during long irradiations. The cross-

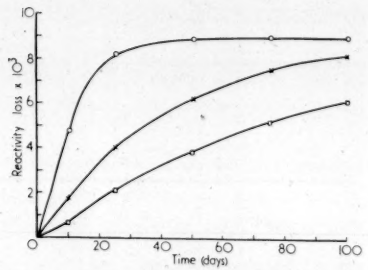

Fig. 8. Effect of samarium poisoning on reactivity during operation

○ $\bar{\phi}$th $= 2{\cdot}6 \times 10^{13}$ n cm^{-2} s^{-1}.
× $\bar{\phi}$th $= 5{\cdot}3 \times 10^{12}$ n cm^{-2} s^{-1}.
□ $\bar{\phi}$th $= 2{\cdot}6 \times 10^{12}$ n cm^{-1} s^{-1}.

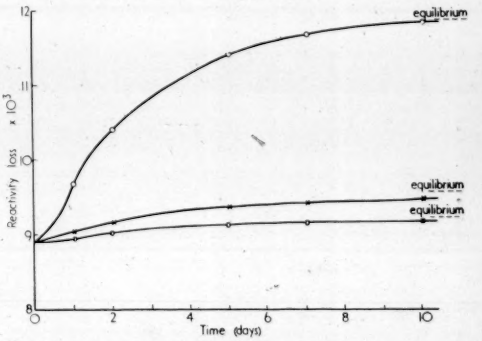

Fig. 9. Effect of samarium poisoning on reactivity after shut down

□ $\bar{\phi}$th $= 2{\cdot}6 \times 10^{12}$ n cm^{-2} s^{-1}.
× $\bar{\phi}$th $= 5{\cdot}3 \times 10^{12}$ n cm^{-2} s^{-1}.
○ $\bar{\phi}$th $= 2{\cdot}6 \times 10^{13}$ n cm^{-2} s^{-1}.

sections of the main isotopes are known reasonably accurately, the overall fission product cross-sections may be subject to an error of $\pm 30\%$. The values of ν are difficult to measure. At present they are not known to an accuracy of much better than $\pm 2\%$ for the pertinent isotopes. The result is that estimated values of the reactivity of a reactor, which depend sensitively on the data used, are subject to large possible errors when one is considering reasonably long burn-up times, of say 3000 MWD/T.

In the case of a natural uranium, graphite moderated reactor it appears that the reactivity will initially fall rapidly due to the formation of ^{135}Xe followed by a further gradual loss due to the build-up of ^{149}Sm. The fission of ^{239}Pu will then lead to an increase in reactivity, greater than the loss due to ^{149}Sm. The formation of ^{240}Pu with its large absorption cross-section then leads to a further continuing fall in reactivity whose rate will be reduced somewhat by the fission of ^{241}Pu. After very long irradiation times ^{242}Pu will begin to build-up. One method of attack on this reactivity problem is to determine, experimentally from irradiated fuel elements, combinations of the nuclear constants which allow long-term predictions to be made.[12]

In a highly enriched reactor the situation will be somewhat simpler as the conversion factor will be small, see Table 8. The main effects will then be a reduction in the concentration of fissile material and the accumulation of fission products. With moderate enrichments the situation will be between these two extreme cases. However, in all cases uncertainty in expectations will be aggravated by repositioning and recycling of the fuel.[13]

In fast reactors a similar state of affairs will exist except that the poisons will all have effects of the same order of magnitude and that more possible fissile isotopes will have to be considered. Further the absorption cross-section of a nucleus may be modified greatly by the scattering cross-section in its effects on reactivity. What is ideally required here is a measurement of an effective cross-section, including the effects of absorption and moderation, as they effect reactivity for the particular reactor. The reactor ZEPHYR, at A.E.R.E., has been used to determine some of the data necessary for an initial assessment of the long-term reactivity changes in a fast power reactor.[14]

Temperature effects. When the temperature of a reactor changes the overall reactivity will usually change. It is desirable that a reactor should have a negative temperature coefficient as it will then be stable to temperature changes. This change in reactivity with variation of temperature is a result of two effects, changes in the density of materials throughout the reactor and changes in the microscopic cross-sections as the neutron spectrum changes.

The density change will affect the macroscopic cross-sections of the materials and also, slightly, the actual size of the system. These may, in turn, affect the reactivity through either, or both, the leakage and k_∞ for the system. The materials will include, in the general case, the fuel, conversion products and the fission products as well as the fuel element sheaths, coolant and any moderator present. The partial reactivity coefficient due to the physical size of the reactor will usually be small and positive. The effect of the changes in macroscopic cross-sections will depend on the actual design of the reactor. Some types of reactor, for example those moderated by water, can be designed so that they have either a positive or negative partial temperature coefficient for this effect, depending on the ratio of fissile material to moderator. It will usually be regarded as important to ensure that the design has a negative partial coefficient in this respect as this will tend to produce a negative overall temperature coefficient.

The partial temperature coefficient due to the neutron spectrum change will also affect both the leakage and k_∞. The effect will depend on the materials present and thus on the previous history of the reactor, also it will be affected by the reactor design as regards the initial materials used. This partial coefficient may vary markedly with the operating temperature of a thermal reactor. In intermediate and fast reactors this partial coefficient will be small as the spectrum is largely independent of the temperature of the reactor.

The overall temperature coefficients of various reactors are shown in Table 10. It will be observed that the range is considerable and the coefficient is negative for all the reactors. In most research reactors the reactivity loss, due to the temperature coefficient, in rising to operating power will be small as the temperature rise will be small. However, in a power reactor where the temperature range between shut-down and full power will be hundreds of degrees the reactivity loss may be large, correspondingly the reactivity gain from operating power to shut-down will be large and will have to be accommodated by the control units. The expected changes may be subject, at present, to some uncertainty if calculated, especially for thermal reactors after long irradiations of the fuel and also for reactors operating just above the thermal neutron spectrum range.

A negative temperature coefficient has been used directly in the CP5 reactor to gain reactivity.[21] Specially chilled water is sent through the secondary side of the reactor heat-exchanger, thus reducing the temperature of the primary coolant and hence increasing reactivity. The procedure is mainly useful for extending the period of time during which the reactor can be sub-critical without the growth of ^{135}Xe prohibiting operation.

There will be a time-lag between a direct change in reactivity which will in general produce a temperature change and the reactivity associated with this temperature change. As a result of such thermal delays the response of the reactor may be such that the neutron density oscillates after a change in reactivity.[15] The thermal time-lag will vary with the reactor design, for example moderator heating will be relatively slow in a gas-cooled graphite reactor compared with that in a water cooled and moderated reactor. If the resultant oscillations are heavily damped out then the reactor

Table 10. *Typical overall reactivity temperature coefficients*

Reactor type	*Coefficient* $\times 10^5$ *per* °C	*Temperature range* °C	*Comments*
Gas-cooled, graphite	$-2{\cdot}8$		BEPO[15]
Water cooled and moderated, research	-8	26–55	B.S.R.[16] varied from -6×10^{-5} to -10^{-4} over temperature range
Pressurized water	-20	~270	PWR, Shippingport[17]
Boiling water	-5 to -22	27–182	As in PWR depends markedly on design[18]
Aqueous homogeneous	-360	~280	Design study, D_2O, circulating fuel[19]
Fast, ^{235}U fuel	$-3{\cdot}5$	38–200	E.B.R.[20]

is said to have good dynamic stability. In the design of a reactor it will be usual to attempt to make the dynamic stability optimum as well as to provide a negative temperature coefficient. In this respect if the fuel itself, or materials in intimate contact with it, are responsible for large negative temperature coefficients then the time-lags will be reduced and dynamic stability increased. Homogeneous reactors and water cooled and moderated reactors where the partial density coefficients can be large and the time-lags small have this advantage. In the Homogeneous Reactor Test, an experimental aqueous homogeneous reactor scheduled for initial operation in 1956, the usual fine and coarse control rods are omitted and replaced by a fuel concentration system which controls the mean temperature of the reactor through the temperature coefficient. The power level is controlled similarly by the rate of heat removal from the reactor. The safety of the reactor against large changes in reactivity is afforded by the temperature coefficient only, safety rods being discarded.[22]

The density effect becomes particularly important in a boiling water reactor where a considerable portion of the core may be occupied by steam. A void coefficient is defined, in this case, as the fractional change in reactivity divided by the fraction of water volume occupied by steam. It has been suggested that the condition be imposed, when designing a boiling water reactor, that the void coefficient should never be positive. However, experience with boiling water reactors indicate that the reactivity taken up by the voids should not be too large otherwise oscillations in the power level occur and that for a high power density a low void coefficient is required.[18] The designed void coefficient for a boiling water reactor will thus be a compromise between power requirements and inherent self-limitation in the event of a power excursion.

The built-in reactivity. In order that the reactor can be raised to a desired power output, maintained there under the envisaged operating conditions and, perhaps, started up again just after shut-down, the maximum value of k_{eff} may be appreciably greater than unity. Thus the possible reactivity may be large; this maximum reactivity for the envisaged operating conditions is termed the built-in reactivity of the reactor. During operation the control system must be capable of accommodating at all times the difference between the built-in reactivity and the required value of the reactivity.

In an enriched fuel research reactor where a reasonable latitude in core design and size is possible, the core will usually be designed so that it provides the required amount of built-in reactivity. Even in this case it may not always be wise to provide all the built-in reactivity which a consideration of all possible operating conditions would suggest. In a large power reactor the limitations on core design and size may be appreciable, the built-in reactivity may then have to take a second place to other considerations such as the mechanical limitation on the size of the reactor pressure vessel. In such a case the operation of the reactor will be planned so that it supplements the available built-in reactivity.

The built-in reactivity which has to be controlled may vary between 3×10^{-3} for a small research reactor to about 0·2 for a high-flux reactor, such as DIDO. A large power reactor of the PIPPA type will probably have a built-in reactivity of between 0·03 and 0·05.

Typical values of the components of the built-in reactivity for various reactors are shown in Table 11. In a research reactor all the built-in reactivity necessary for full operation may not be made available if the reactor is to be used for limited operation only. The values in Table 11 are estimated from published data on the operation of the Materials Testing Reactor[23] and the Experimental Boiling Water Reactor[24]; in the case of the other two power reactors they are possible components denoting the order of magnitude only. The control system will have to accommodate all this reactivity; the actual control investment available will be larger than this to take into account errors in estimates of the built-in reactivity, to provide as rapid a shut-down as is required and to ensure an adequate safety factor in the case of shut-down. There will be a tendency to make the built-in reactivity as small as possible so that the control investment is reduced, thus reducing the risk of a damaging power transient owing to misuse of the control system; and possibly decreasing the capital cost of the reactor.

Table 11. *Typical built-in reactivities and their components*

Reactor	*MTR*	*EBWR*	*Fast*	*Gas cooled graphite*
ϕth	2×10^{14}	$1{\cdot}6 \times 10^{13}$	—	10^{13} $cm^{-2}\,s^{-1}$
Power	30	20	500	500 MW
Operating temperature	76	252	500	350 °C
Purpose	Irradiation facility	Power prototype	Power	Power
Built-in reactivity	18·9	9·3	1·35	$4{\cdot}2 \times 10^{-2}$
Components $\times 10^2$				
^{135}Xe	3·9	3·36	0	2·5
^{149}Sm	1		0	0·4
Depletion, and other fission products	7	—	0	
Temperature	0·8	3·5 + 2·2*	1·25	1·2
Experiments	1·5	—	0	0
Operations	4·7 (xenon) (over-ride)	0·23 (xenon) (over-ride)	0·1	0·1

* 17·5% steam voids.

CONSIDERATIONS OF SAFETY

A reactor which has been operating at power for some time contains the radioactivity associated with the fission products. The activity of these products in a 500 MW reactor after a reasonable period of operation amounts to about 600 Mc. It has been suggested that if 0·1% of the fission products escaped from such a reactor, with reasonable atmospheric conditions, evacuation of the population, or several restric-

tions on normal habits of living, would be necessary, downwind, to a distance of about two and a half miles.[25] It is essential that the possibility of escape of fission products should be kept to the minimum by careful design and operation of a reactor. The problem is aggravated as the power may rise rapidly if maloperation occurs. For example if a reactivity step of 10^{-2} is inserted into a critical system the time taken for the power to double is 20 ms with a water cooled and moderated reactor, and about 35 μs in the case of a fast reactor. In practice a step change in reactivity cannot be applied so that the actual "doubling times" will be somewhat longer than these; if the reactivity is added at a rate of $10^{-2}\,s^{-1}$ to the water cooled and moderated reactor then the doubling time is 0·35 s, still quite a short time.

The safety of reactors is a large and vital subject and only a few points pertinent to the control system can be mentioned here. The electrical[26] and mechanical[27] engineering considerations which arise have been described recently. In general safe operation will be achieved by a combination of:

(*a*) designing the reactor so that the possibility of an accident is minimized or so that it is inherently safe,
(*b*) strict operational procedure,
(*c*) designing the control system so that under all conditions it gives maximum protection to the reactor.

The first item indicates that the built-in reactivity must be as small as possible so that the reactivity absorbed by the control rods is a minimum. The possibility of a damaging transient will then be reduced if the control units are misused. Similarly the items absorbing reactivity, whether they be control units or parts of an experiment, should individually absorb as little as possible. A strong negative temperature coefficient, especially with small thermal time-lags, will further promote the safety of the reactor. If the fission products are continuously removed from the reactor then the problem of safety changes location from the reactor to the associated chemical plant. This procedure will have another advantage in that the xenon poisoning will not build up and so both the built-in reactivity can be reduced and, from the control view point, more latitude in the operational time scale will be possible. The inclusion of fuses, operated as directly as possible by the neutron flux, will be an added safety factor.

The fundamental safety principles on which the control system design will be based are:

(1) Rates of addition of reactivity will have a set maximum value.
(2) The shut-off units should always be able to reduce reactivity rapidly.
(3) The system must "fail safe," i.e. whatever fault occurs in the system it must tend to produce a safe condition of the reactor; it should sound an alarm or start to make the reactor sub-critical.
(4) The neutron density must be monitored at all times except when the reactor is fully shut-down. This is true especially when the reactor is being taken to the critical state.
(5) Reactor power should be monitored by at least two independent monitors, preferably of different types.
(6) Ease of operation and smoothness of control of reactor power by the system will be a major safety factor.

Another positive safety factor will be operation of the reactor by a competent and well-organized team.

CONCLUSIONS

The general conclusion may be drawn that no large unforeseen fundamental problems are likely to arise in the future concerning the control of reactors. Analogue and digital computors will be used increasingly, with a resulting general refinement of methods and techniques of calculation, for solving the known problems of control.

However, the best source of information on the operational and control features of any given reactor type, especially when the reactor has to provide useful power, is, and will continue to be, direct experimental evidence from a prototype reactor. Other considerations aside, this will be a strong reason for building a pilot reactor, from which the optimum information regarding control can be obtained, before embarking on the building of a full-power reactor.

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APPENDIX

List of symbols

a = A form factor.
C_i = Number of atoms of precursor of ith delayed neutron group in reactor.
E_n = Mean neutron energy.
E_0 = Mean fission neutron energy.
E_{th} = Mean thermal neutron energy.
f = Thermal utilization factor.
i = Subscript denoting a delayed neutron group.
k_{eff} = Effective reproduction constant.
k_{ex} = Excess reproduction constant = $k_{eff} - 1$.
k_∞ = Reproduction constant of system of infinite size.
l = Thermal diffusion time, s.
l_s = Slowing down time, s.
n = Neutron density in reactor.
P = Thermal power output.
P_D = Delayed thermal power output.
P_P = Prompt thermal power output.
S = Neutron source strength, neutrons/s.
t = Time, s.
T = Reactor period, s.
T_0 = Reactor operating time, s.
v = Neutron velocity.
V = Volume of reactor core, cm^3.
β = Total fractional yield of delayed neutrons, as fraction of total neutrons per fission.
β_i = Fractional yield of ith delayed neutron group.
γ = A fission product yield as a fraction of total fission product yield for thermal fission of ^{235}U.
λ = Mean decay constant of delayed neutrons.
λ_i = Decay constant for ith delayed neutron group.
ν = Number of neutrons produced per fission.
ρ = Reactivity of system = $(k_{eff} - 1)/k_{eff}$.
σ_a = Microscopic absorption cross-section, cm^2.
τ = Mean prompt neutron lifetime, s.
$\tau_{\frac{1}{2}}$ = Half-life for decay of radioactive isotope.
$\overline{\theta_{th}}$ = Mean thermal neutron flux, n cm^{-2} s^{-1}.
ω = Parameter having dimensions of inverse time, s^{-1}.

The instrumentation of reactors

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The paper presents a survey of the instruments needed for the normal operation of reactors. All reactors produce power: the instruments used for its measurement and control and its effect on the temperature of the reactors components are described. These include instruments operated from detectors sensitive to neutron flux and those concerned with physical measurements such as temperature and flow of coolant. The principles of operation of the various instruments are described and their relative importance under various operating conditions indicated.

In power reactors instruments are also necessary for the determination of the change of the neutron flux pattern with time and for the detection of faults in protective fuel containers. The basic principles of such instruments are described. An assessment of the essential instruments for a large power producing reactor is made.

1.0 INTRODUCTION

Reactor instrumentation in its fullest sense covers a very wide field of operational and experimental instruments but here only instruments necessary for the safe and continuous operation of reactors are discussed. All reactors from so-called "zero energy" experimental to power reactors rated at hundreds of megawatts are power producers and it is with the measurement and control of this power and its effect on the temperature of the reactor components and coolant that most of the instruments are connected. The actual problem of control is dealt with in a complementary paper.[1] The instruments described are part of the means of effecting control. There are two main groups associated with power measurements, those operated from detectors sensitive to neutron flux—flux measuring instruments—and those concerned with physical measurements such as temperature and coolant flow.

In low power experimental reactors and at the start up of power reactors, the power produced is too small to be measured by calorimetric methods. Instead it has to be deduced from the measurement of neutron flux to which the power is related. When the power output is sufficient to raise the temperature of the fuel and coolant significantly calorimetric methods can be used for the accurate measurement of the power but because of their rapid response, flux measuring instruments are still valuable for detection of changes in power level and for the operation of safety circuits.

In addition to instruments associated with power, two other types of instruments are required for power reactors. Because the nuclear properties of the fuel are a function of irradiation, the neutron flux pattern in the core changes with time. This could cause local overheating, or if this were checked, a reduction in output unless adjustments are made. The latter can only be carried out after changes have been shown up by instruments which can measure the flux distribution in the core. When the fuel is incompatible chemically with the coolant, it is vital that faults in the fuel containers are detected rapidly if damage or serious contamination of the reactor and coolant are to be avoided. To ensure early warning of such failures elaborate equipment, which can be an appreciable fraction of the whole instrumentation, is necessary. The basic principles of the instruments used for flux distribution measurements and for detection of faulty fuel containers are discussed.

2.0 FLUX MEASURING INSTRUMENTS

2.1 POWER MEASUREMENTS

The neutron flux in any region in a reactor core is proportional to the fission rate and hence to the energy released in that region. The flux is greatest at the centre of the core and decreases to a minimum at the surface (from which many neutrons escape) and the energy release is likewise distributed. The total power output is the integrated power over the whole core. Fortunately, to a first approximation, the flux pattern throughout the core is independent of the total power so that measurement of flux at one position in the core can be used as an indication of the operating power level. Similarly, the pattern of neutrons escaping from the core is unaffected by the power level and measurement of flux at a point adjacent to the core can be used as a measure of total power. It is convenient in most reactors to measure the flux adjacent to and not in the core except during start up conditions when greater sensitivity may be needed.

It is essential for safe operation of a reactor that the power level and rate of change of power level be indicated at all times by instruments with rapid response. This is particularly so at start up when the reactor period may become very short causing the power to increase rapidly. Unless this is detected immediately and action taken, a dangerous condition could result. Flux measuring instruments are suitable for this duty.

Unfortunately because of the large range of power that has to be covered from the subcritical to maximum power, no single instrument or one type of instrument can cover the range. For example, the power of a 100 MW thermal reactor when shut down could be about 1 milliwatt. In this event the instruments would have to measure power over 11 decades. In practice it is usual to raise the minimum power of a reactor by a factor of 100 or 1000 by insertion of neutron sources into the core, but even so the power range to be covered is considerable.[2]

There are two basic types of flux measuring instruments, those which utilize the direct current output of an ionization chamber and those which count the pulses from a fission chamber. The former are simpler and more versatile, the latter more sensitive. It is usually necessary to employ some instruments of both types in order to cover the power range adequately.

2.1.1 *Detectors*

Ionization chambers. An ionization chamber can be used to detect fast or thermal neutrons. When detecting the former the chamber is filled with hydrogen or a hydrogen compound. Protons of high energy released by the collision of the fast neutrons and hydrogen atoms caused ionization of the gas and hence a current to flow through the chamber proportional to the neutron density. Such a chamber has a sensitivity to gamma-rays of the same order as to fast neutrons and as gamma-rays of high intensity are also emitted in the fission process in the reactor some form of

discrimination against gamma-rays has to be employed. This can be done by the use of two chambers "back to back," one of which is made sensitive to gamma-rays only by filling with another gas such as argon. However, in practice only partial compensation can be achieved and for power measurements it is preferable to slow the neutrons down in a moderator and detect them by chambers sensitive to slow neutrons.

One design of ion chambers is shown in Fig. 1 described by Abson and Wade[2] and is a development of the original design by Carmichael. The outer case and the electrodes are

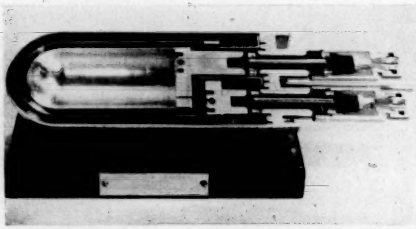

Fig. 1. Section of type RC1 ionization chamber

of high purity aluminium. The latter are supported on quartz insulators and steel bolts which also act as terminals. This chamber can be filled with boron trifluoride gas but better voltage saturation characteristics are obtained with boron coated electrodes and hydrogen filling. The sensitivity is proportional to the area of coated electrode but the saturation is improved if the coating is restricted to the cylindrical portions of the electrodes where the electric field is uniform. A further improvement in the saturation characteristic is obtained if only the negative centre electrode is coated.

The sensitivity is also a function of filling pressure. It increases in a non-linear manner with pressure up to about two atmospheres above which little improvement can be achieved. The efficiency of collection of the ionized gas increases with decrease in pressure since the ionization density is reduced and the speed of collection increased. Thus the polarizing voltage required to give satisfactory saturation decreases with decrease in pressure. While the intrinsic sensitivity is unaffected by temperature the collection characteristic is impaired. To overcome this and to ensure that the sensitivity is unaffected by small changes in polarizing voltage it is usual to make the latter two to three times the nominal saturation voltage. For example, a coated chamber filled to two atmospheres, while saturating at 200 V in a flux of 10^9 n cm^{-2} s^{-1}, should be polarized by a 600 V supply.

The action of the chamber depends upon the nα reaction of the B^{10} isotope with slow neutrons.

$$B^{10} + n^1 \rightarrow Li^7 + He^4$$

Boron rich in the B^{10} isotope permits the sensitivity of a chamber to be increased by a factor of 5 over that when natural boron containing only some 18% of the B^{10} isotope is used.

Thus where the greatest sensitivity is needed, i.e. when the flux to be measured is low, as much electrode area as possible should be coated with the B^{10} isotope and a filling pressure of two atmospheres should be used. The best sensitivity that can be achieved is approximately 3×10^{-14} A/n cm^{-2} s^{-1}.

The ionization of the filling gas by gamma-rays is directly proportional to the gas pressure and as the neutron sensitivity/pressure characteristic tends to saturate at higher pressures, the highest neutron/gamma sensitivity ratio is obtained with a low filling pressure when unfortunately the neutron sensitivity is least. It follows that with a given polarizing voltage, the best discrimination against gamma interference is obtained with a low filling pressure. At the same time the maximum operating flux increases with the decrease in filling pressure because of the better collection characteristics. With the RC1 type chamber coated over 500 cm^2 with natural boron and filled with hydrogen at a pressure of 1/5 atmosphere, the neutron/gamma sensitivity ratio is 400 n cm^{-2} s^{-1}/r h^{-1}. With enriched boron the ratio could be reduced to 80. When greater discrimination is required or when the loss in neutron sensitivity due to reducing the filling pressure cannot be tolerated, an external gamma shield has to be used. Pure lead affords good gamma shielding with relatively low neutron absorption and can improve the neutron/gamma sensitivity by at least 100 fold.

When exposed to radiation the materials of the chamber become active and emit beta and gamma rays which give rise to interference. Under steady conditions this may not be serious but when the flux level is reduced rapidly, such as when a reactor is shut down, the chamber current due to this activity can swamp that due to neutrons. The main activity, which is from the aluminium, is short lived and falls off 1000 fold in half an hour after removal from the neutron flux. The remaining activity from impurities and electrode leads dies away more slowly and can prevent the chamber from indicating the lowest neutron flux for some hours. Normally the range of reactor power is covered by at least two ion chambers and in order to reduce the effect of this source of interference it is usual to remove the chamber covering the low power range from the measuring position while the reactor is running at high power.

Fission chambers. A typical fission counter chamber is shown in Fig. 2. It consists of a stainless steel containing

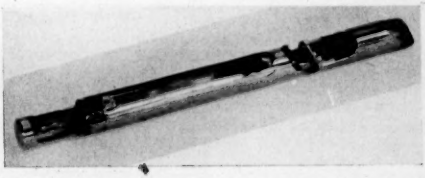

Fig. 2. Section of a fission chamber

tube concentric with, and insulated from which, is a cylinder coated with uranium rich in the ^{235}U isotope. The cylinder is held at a negative potential with respect to the container. Neutrons cause fission of the uranium and the fission fragments ionize the filling gas and cause pulses of current to flow through the chamber. The coating of uranium is limited to about 1 mg/cm^2. With thicker coatings the chance of fragments of atoms fissioned near the cylinder escaping and ionizing the gas is reduced. For a constant coating thickness the sensitivity increases with the coated area. For the type of chamber described the sensitivity is about 0·0005 counts s^{-1} cm^{-2} in a flux of 1 n cm^{-2} s^{-1}, i.e. with a coated area of 100 cm^2 the output is 0·05 pulses a second. The pulse is of

the order of 0·1 μs and it is possible to count at rates up to 5×10^5 c.p.s.

The pulses produced by the direct ionization of the filling gas by gamma rays are lower in amplitude than those resulting from fission and it is possible to discriminate against them in an amplifier circuit. This permits the chamber to operate satisfactorily as a neutron detector in gamma intensities as high as 10^5 r/h.

A fission chamber can detect fast neutrons but as the fission cross-section of uranium for fast neutrons is much smaller than for thermal neutrons, the sensitivity of the chamber is considerably less than when detecting the latter.

Boron trifluoride proportional counter. A typical boron trifluoride counter consisting of a fine wire concentric with, and insulated from, a thin copper tube is shown in Fig. 3.

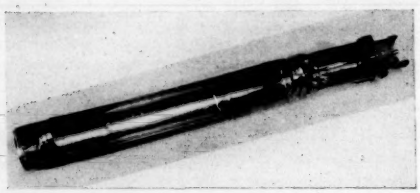

Fig. 3. Section of a boron trifluoride proportional counter chamber

Fission of boron atoms of the enriched boron trifluoride filling causes ionization of the gas and pulses of current to flow across the chamber. Proportional counters are about 100 times more sensitive to slow neutrons than fission chambers of similar size, but unfortunately their gamma sensitivity is also considerably greater and it is not possible to discriminate between gamma and neutron pulses which are of similar amplitude. The duration of the pulses is longer than those from fission chambers and this reduces the maximum permissible counting rate to 10^4 c/s.

Other neutron detectors. Thermopiles consisting of a number of thermocouples in series, the junctions being coated with boron have been used as neutron detectors.[3] The energy released by fission of the B^{10} isotope by thermal neutrons heats up the coating and a potential is developed across the thermopile. Uncoated thermo-junctions in series opposition to the neutron sensitive ones can be used to at least partially compensate for ambient temperature changes. Thermopiles are relatively slow in response compared to ion or fission chambers and their sensitivity is affected by temperature. These characteristics make them unsuitable for power measurements. Their main use as probes when exploring the flux distribution in experimental cores is described in Section 2.2.

2.1.2 *Measuring circuits using ionization chambers*

Ion chambers, because they give a direct current output, require simpler amplifying and indicating equipment than fission chambers and proportional counters and are used for power measurement whenever possible. Their usefulness is restricted at lower flux levels by the minimum current that it is possible to amplify and in some situations by gamma interference.

The simplest circuit is that shown in Fig. 4(*a*). The ion chamber is polarized by a battery and the output current flows through a galvanometer. Shunts across the latter enable the measuring range to be varied. An ion chamber coated with boron over 500 cm^2 in a flux of 10^{10} n cm^{-2} s^{-1} could provide a current of 50–60 micro amperes, so that with a meter with a full-scale deflexion of 1 micro ampere, power could be conveniently measured over a range of three decades. The range could be extended by using a more sensitive meter

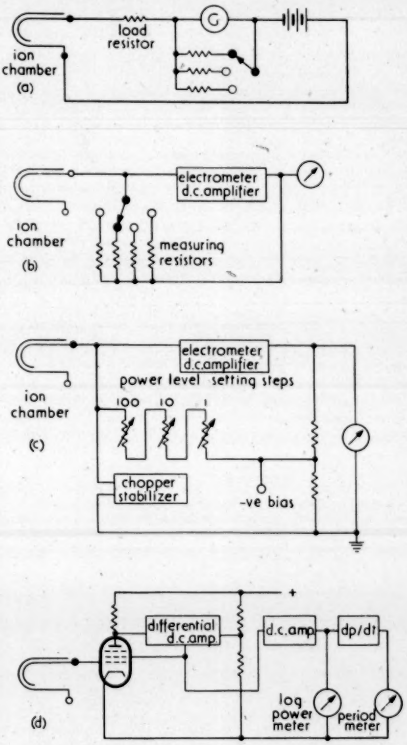

Fig. 4. Measuring channels using ionization chambers

or better by moving the ion chamber into a region of higher flux when at the lower powers. This circuit has the advantage that it does not depend upon any external supplies and can therefore provide indication at all times. It is, however, not very suitable for operation of alarm and safety circuits.

In the second circuit, in Fig. 4(*b*), the galvanometer is replaced by an electrometer amplifier. The output voltage of the amplifier balances the voltage developed across the measuring resistor. By switching this resistor the range can be varied over a wide range. However, to reduce the effect of drift in the amplifier a full-scale output voltage of 10 V is required and this involves the use of measuring resistors of

up to $10^{12}\ \Omega$. The accuracy and stability of these resistors determine the accuracy of the measurement and as precision wirewound resistors can only be obtained economically up to $10^{7}\ \Omega$, a fall off in performance has to be accepted in the lower power (higher resistance) ranges for which carbon resistors have to be used. This limitation is not too serious so long as it only occurs in ranges three or more decades below normal operating power levels.

The third circuit shown, in Fig. 4(*c*), is one used to indicate changes in reactor power from a preset level. Having detected the deviation, it can provide an initiating signal for appropriate operation of the control rods to correct the drift in power. The amplifier used is similar to that of the previous circuit but a higher degree of stability is required if the circuit is to be used to maintain the reactor power within close limits. It is not unreasonable to require that the power be held within $\pm 1\%$ in which case the stability of the measuring circuit should be one order better, i.e. 0·1%. To achieve this a chopper type stabilizer is employed. It can be so effective in reducing the drift that the voltage across the measuring resistors can be reduced so that 100 mV corresponding to full output of the amplifier is still appreciably greater than an equivalent drift signal referred to in the input. The value of the measuring resistors are reduced proportionally with the voltage and it is possible to use wirewound resistors for all ranges. As the circuit is generally required to operate alarms and control circuits it is desirable for it to have a reasonably high output voltage. This requirement and that of low voltage across the measuring resistors are met by deriving the latter from a voltage divider across the amplifier output and biasing it with a negative voltage.

The fourth circuit is one in which the output voltage varies as the logarithm of the current from the ion chamber. Because of this characteristic it can indicate power over a wide range which is of particular value while starting up a reactor. When the power of a reactor is increasing it does so exponentially; the ion chamber current does likewise and the output of the logarithmic amplifier increases linearly. The rate of rise of this voltage proportional to reactor power is the inverse of reactor period and can be measured by a differentiating circuit and displayed on a meter suitably scaled in terms of reactor period.

The heart of the circuit is the logarithmic part of the amplifier. Early experiments with thermionic diodes showed that a diode operated in the retarded field region exhibited an exponential current/voltage relationship over a considerable range.

$$I = I_0 e^{rj/kt} \qquad (1)$$

The list of symbols will be found in the appendix.

If a diode operating under such conditions replaces the measuring resistors in the electrometer valve amplifier of circuit in Fig. 4(*b*) the output voltage varies as the logarithmic of the input current from the ion chamber. The above relationship shows that the current flowing in the diode is determined by the absolute temperature of its cathode as well as the voltage across it. The practical difficulty in using the diode in the circuit is maintaining the temperature constant in order to eliminate the temperature effect or to correct for it in some way. The obvious first step is to stabilize the heater voltage as carefully as possible. Partial compensation can be achieved by using the temperature drift signal from a second diode to balance that of the first. However, the temperature characteristics of two apparently similar diodes are rarely identical and the gain that can be obtained by such a scheme is limited to a factor of about 10.

An alternative method of compensation has been developed and is now used almost exclusively in this type of amplifier in this country.[4] The diode is replaced by a suitable pentode valve as shown in Fig. 4(*d*). A differential amplifier working between the anode of the pentode and a reference voltage provides the screen voltage. Changes in anode and screen currents which would result from changes in ion chamber current are prevented by the automatic adjustment of the screen voltage. The screen/cathode characteristic of the valve is effectively that of the grid/cathode characteristic multiplied by a factor and so that the screen voltage varies logarithmically with the current from the ion chamber. Now any change in effective grid/cathode potential caused by temperature variations which would cause the anode and screen currents to change are counter balanced by a proportional change in screen voltage. With such an arrangement it is possible for the logarithmic amplifier to operate over six decades. The signal to the period indicator is the inverse of the period, one end of the scale corresponding to infinite period and the other to some design value usually about 20 s.

When a reactor is up to working temperature and in the "power range"—say above 1% of full power—it would be

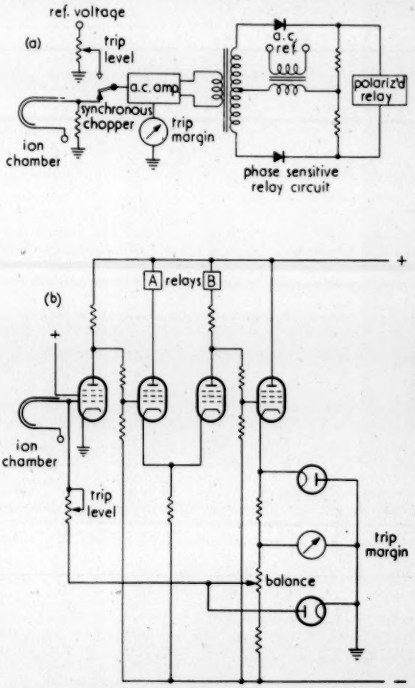

Fig. 5. Safety trip circuits operated from ionization chambers

dangerous to allow the period to fall as low as 20 s, 2000 s or more being more acceptable. Clearly a safety device operated from a circuit measuring such long periods would be too slow to be of use. Instead, measuring channels which detect small changes in power from a preset level are used to give a tripping or alarm signal. They provide the same information as the power drift circuit of Fig. 4(*c*) but they are designed as safety devices rather than indicators and incorporate features to ensure that as far as possible they "fail safe" and so do not jeopardize the reactor. Two types have been developed in this country,[5,6] an a.c. and a d.c. type which are shown schematically in Figs. 5(*a*) and 5(*b*) respectively.

In the a.c. version the ion chamber signal is compared to a reference voltage by means of a precision synchronous chopper device. The difference voltage is amplified and fed to a phase sensitive circuit and holds in a polarized relay. When the voltage across the ion chamber load resistor equals the reference voltage, the input to the amplifier falls to zero and the relay is de-energized. The phase sensitive circuit prevents the relay from being re-energized should the ion chamber signal exceed the reference voltage.

In the d.c. version the ion chamber feeds an electrometer valve which is connected to the grid of one of two cathode coupled pentode valves. In the anode circuit of each of these valves is a relay. The bias on the grid of the second valve of the pair is such that in the quiescent state both valves take similar currents. The anode of the second controls a cathode follower which provides the feedback signal. When the ion chamber current equals a preset value determined by the size of the feedback resistor, the cathode potential of the cathode follower has fallen to earth potential where it is held by a diode. The balance of the circuit is then upset as the feedback has been removed. The first valve of the coupled pair cuts off and relay *A* is de-energized. Too great a decrease in ion chamber current or any failure in the circuit which causes excessive rise in the cathode potential of the cathode follower is detected when the tapping point for the feedback rises to earth potentials where it is held by a second diode. Again the feedback is removed and the second valve of the coupled pair cuts off and the second relay *B* de-energized.

2.1.3 *Measuring channels using pulse counters*

The instrument requirements for fission chambers and boron filled proportional counters are very similar. The simplest equipment consists of the counter chamber, a head amplifier mounted as close to the chamber as possible, a main amplifier, a pulse discriminator and a scaler. The scaler counts and indicates the pulses received from the chamber during a given time interval.

Such a channel is useful for very low count rates such as obtain during initial loading of fuel into the reactor, but it is more convenient to have the information presented as a continuous meter reading. This can be provided by a ratemeter fed from the scaler. The ratemeter can be a linear device with a number of ranges of logarithmic.[7] The latter version can operate over six decades and, like the logarithmic amplifiers following an ion chamber, can provide a signal proportional to the inverse of reactor period.

Measuring channels using pulse chambers are used normally in reactor power measurement at the lower end of the power range at which it may not be possible to use ion chambers. The choice of chamber, fission or boron trifluoride proportional counter depends upon conditions. The proportional counter, being the most sensitive, permits lower powers to be measured in a given measuring position provided the gamma background is not too high. The fission chamber can cover a greater range and has better discrimination against gamma radiation. A practical advantage of the fission chamber is that it requires a polarizing voltage of about 500 V against 1–2 kV needed by the proportional counter.

2.2 FLUX SCANNING

While the neutron flux pattern in a reactor core is sensibly independent of the operating power level, it is a function of irradiation time of the fuel. Many factors influence the change and their effect is not accurately predictable but the main pattern of cause and effect is clear. Neutrons are expended in fresh fuel in fission, or capture useful or otherwise in certain proportions. In time these proportions change. In a thermal reactor using uranium fuel, the first major change is due to a build up of stable fission products which capture neutrons non-productively. This effect is subsequently more than counter-balanced by the formation of ^{239}Pu. The plutonium, which is fissile, and what is left of the ^{235}U, present a greater total cross-section to thermal neutrons than the original amount of ^{235}U and so a greater proportion of the neutrons are used in fission. The proportion of neutrons used in fission increases to a maximum and then falls off progressively in a manner determined by the increase in ^{239}Pu, depletion of ^{235}U and the formation of higher isotopes of plutonium. The effect of these changes in terms of reactivity for a small section of a reactor core is shown in Fig. 6. Now as the neutrons flux varies over the

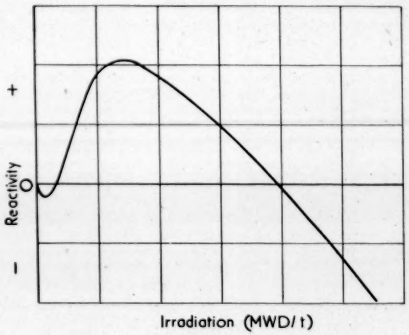

Fig. 6. Variation of reactivity with irradiation

core, being greatest in the centre and decreasing towards the outside, the local changes in reactivity at the centre are more rapid than in outer regions. Because of this, during the plutonium build up, for a given total power output the neutron flux and power output increase at the centre and decrease at the outside. The increase in power from the centre implies higher fuel temperatures which may not be permissible for metallurgical reasons. In this event unless corrective measures are taken, such as inserting absorbing material into or removal of fuel from the centre core, the reactor power has to be reduced so that the safe temperature is not exceeded.

The time scale of this irradiation effect on the flux pattern is proportional to the fuel rating. In a low rated reactor many years elapse before the increase in reactivity due to plutonium build up is reached but in power reactors with fuel ratings of 1 MW/t or more, significant changes are apparent in a matter of weeks. Accurate determination of the flux distribution (i.e. flux scanning) of these reactors in particular is essential. In experimental reactors it is also desirable to be able to measure the flux distribution in order to obtain information on the effect of alterations in the core structure on the flux pattern and also to measure the distortion of the flux pattern by the insertion of experimental apparatus or samples of materials for irradiation.

One method of measurement particularly applicable to experimental reactors is to irradiate small samples of materials such as manganese and iridium at various points in the core and to measure their activity on removal. The "foil" technique, as it is known, though laborious is accurate and, because of its sensitivity, particularly suitable for the measurement of low flux levels. It was mentioned in Section 2.1.1 that thermocouples coated with boron can be used for determination of thermal neutron flux. The output of a couple (or of a number of them connected in series to give a larger output) as it is passed down channels in the reactor core is a measure of the relative flux distribution along the traverse. By traversing a number of suitable channels a picture of the flux pattern can be built up. Absolute values can be given to the results if the couple has first been calibrated against an ion chamber of known sensitivity or against foils. Unfortunately the effect of temperature on the sensitivity of the coated couple makes it unsuitable for use in high temperature power reactors. Small ionization chambers have also been used for flux scanning. Their use is restricted because of structural considerations to about 100° C and by saturation difficulties to flux levels below 10^{11} n cm^{-2} s^{-1}.

A third method which can be regarded as a development of the foil technique, depending on the build of gamma or beta activity of certain materials under irradiation, is the only satisfactory one so far developed which is suitable for high temperature power reactors. The intensity of the emission from a sample after removal from the core is a function of the neutron flux level at the region in the core where the material was located, the time it was in the core and the time subsequent to removal at which the gamma or beta activity is measured. A convenient form for the sampling material is a wire or chain which can be lowered into vertical channels in the core. The gamma or beta activity builds up along the wire in proportion to the neutron flux distribution. After a "soaking" the wire is withdrawn and the activity measured as it is drawn through a detector. For good results the wire material must be such that the time required to induce gamma or beta activity and its decay are long with respect to the time needed to insert it into and remove it from the core. Otherwise the activity of the wire will be appreciably affected by the various regions of the core through which it is passed. The lower end of the wire will be most affected giving a false impression that flux in the bottom end of the core is higher than it really is. The alternative is to shut the reactor down or to run it at low power while inserting or removing the wire, but this, of course, is undesirable with a power reactor.

For the same reasons the decay of the activity must be long with respect to the time needed to pass the wire through the measuring equipment. On the other hand the half-life must not be so long as to require an inconveniently long "soaking" time in order to become sufficiently active. A slow decay would reduce errors due to time in measurement but the wire would remain so active as to preclude its use again for a long time, if ever. Storage of this material could become an embarrassment as time went on. Suitable wire materials from the nuclear viewpoint are copper, manganese, tungsten and platinum. The abundance of the primary isotope, cross-section to thermal neutrons and the half-life of the isotope produced under irradiation are shown in Table 1.

Table 1. *Nuclear characteristics of materials suitable for flux scanning wires*

Primary isotope	*Abundance (%)*	*Absorption cross-section (barns)*	*Radio-active isotope*	*Half-life (h)*	*Gamma and beta emission (MeV)*
^{63}Cu	69	4·3	^{64}Cu	12·8	1·34 (β) 0·66 (γ)
^{55}Mn	100	12·6	^{56}Mn	2·6	0·7–2·9 (β) 0·84–3·0 (γ)
^{186}W	28·7	34	^{187}W	24	0·6, 1·33 (β) 0·5 (γ)
^{196}Pt	25·4	1·2	^{197}Pt	18	0·7 (β) 0·08, 0·19 (γ)

It is important that impurities are closely controlled because even small traces can influence the activity of the wire significantly. Tungsten and copper are favoured because they can be obtained in very pure form at an economic price. Tungsten retains greater mechanical strength at high temperatures than copper and is therefore preferred.

The activity induced in the material in disintegrations per second is given by

$$\text{Activity} = N\sigma_a\phi(1 - e^{-\lambda T}) \qquad (2)$$

Using tungsten in a flux of 10^{13} n cm^{-2} s^{-1} for 1 h

$$\text{Activity} = 1{\cdot}81 \times 10^{22} \times 34 \times 10^{-24} \times 10^{13}(1 - e^{-0{\cdot}0288}) = 1{\cdot}72 \times 10^{11} \text{ dis s}^{-1}\text{ cm}^{-3}$$

This is equivalent to 4·65 c cm^{-3}.

Using 22 s.w.g. wire, the activity per centimetre run would be 18·5 mc. This is the value immediately the wire is removed from the core. Assuming that measurements cannot be made until an hour later, the level will have fallen by just under 3% for which due allowance can be made.

3.0 PHYSICAL MEASUREMENTS

3.1 TEMPERATURE

When any appreciable power is being generated it is essential that a check is kept on the temperature of the fuel, coolant, moderator and surrounding structure. The measurements are normally made using conventional thermocouples, although resistance thermometers are sometimes more convenient for the measurement of coolant temperatures. In a small experimental reactor the measurements involve only a few thermocouples but in a large power reactor temperature measurement may involve up to 1000 couples and associated indicators and recorders. This can be illustrated by considering the temperature measurements desirable in a large gas cooled reactor.

In the core of such a reactor are thousands of channels containing the fuel and through which the coolant is passed.

The heat release is greatest in the channels at the centre of the core and least in the outermost channels. Also in each channel it is greatest at the mid-section and minimum at the ends. Coolant passing into the channel picks up heat at an increasing rate until it reaches the mid-section of a channel and at a diminishing rate thereafter. The maximum temperature difference between fuel and gas is at the mid-section where the heat rate is greatest but further up the channel the still rising gas temperature more than counteracts the fall off in heat rate causing the maximum fuel surface temperature to occur towards the outlet end of the channel, the actual position depending upon the heat transfer characteristics and the pattern of the heat release up the channel.

A typical distribution is shown in Fig. 7. In power reactors in which it is desirable to achieve as high a bulk output gas

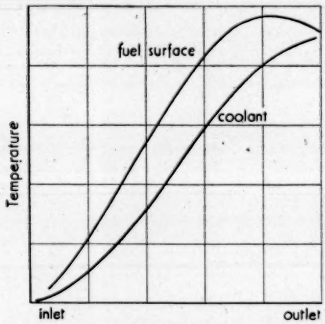

Fig. 7. Fuel surface and coolant temperature distribution along a fuel channel

temperature as possible, the coolant flow through the outer lower rated channels is restricted and in consequence the fuel surface temperature and gas outlet temperatures may be very similar to those of the more highly rated central channels. It is therefore not sufficient to monitor the temperature of the fuel in the central regions of the channels only. Ideally the maximum fuel surface and gas outlet temperature of each channel should be checked but a practical compromise is to monitor about 3–4% of the channels.

To measure gas outlet temperatures metal sheathed mineral insulated thermocouples are used. These are built into the core during construction. The measurement of fuel surface temperature is more difficult. Leads attached to fuel interfere with fuel handling arrangements and much ingenuity has been used in devising ways of overcoming this. Many of them involve making electrical contacts inside the reactor core which tend to make the measurement less reliable. However inconvenient, thermocouples can be attached to the fuel. It may not be possible to place the thermocouple at the point of maximum temperature, but, as shown in Fig. 7, the temperature difference between the point of maximum temperature and the channel outlet is small and from a measurement at the latter the maximum temperature can be estimated. The temperature of the core material has also to be monitored. Thermocouples are embedded in it in a pattern such that the radial and axial temperature gradients can be determined. As these couples cannot be replaced, enough spare couples have to be inserted to give a fair assurance that the measurement can be made throughout the life of the reactor. In a power reactor these thermocouples measuring fuel coolant and core temperature have to be brought out of the containing vessel through seals capable of withstanding elevated pressures and temperatures.

In addition to radiation and conduction from the core, the containing vessel and biological shielding are heated by absorption of neutrons and gamma rays. A check must be kept on their temperature and temperature gradients in them by a large number of thermocouples permanently embedded in them to ensure that they are not being overheated or overstressed. Allowing for spare capacity some 500 couples may be required.

The primary function of temperature measurements so far discussed is that of safety. For normal measurements of power and for control purposes, thermocouples or resistance thermometers are needed to measure the temperature of the bulk coolant at the inlet to and outlet from the core. These detectors must be designed and sited so that their response to any change in conditions in the core or in external apparatus is as rapid as possible.

At least some of the vital temperatures such as fuel element and channel coolant temperatures must be continually indicated and recorded. The others need only be checked automatically or manually at intervals. For both purposes standard commercial instruments are satisfactory.

3.2 COOLANT FLOW

To measure the flow of gas or liquid coolants through reactor cores orthodox flow meters operated from venturi or orifice plates in the coolant circuit are used almost exclusively. An exception is the use of magnetic induction devices for liquid metals. These have, however, to be calibrated by the orthodox devices.

Of the energy expended on forcing the coolant around the circuit in experimental reactors of appreciable power and in power producing reactors, only part can be recovered and so it is economically desirable for this pumping power to be as low as possible. Unfortunately the rating of nuclear fuel in power reactors is so high in relation to the heat transfer surface that can be provided that in order to keep the fuel surfaces temperature within prescribed limits, at present in the region of 500° C, high coolant velocities have to be employed in the coolant channels with consequently high pressure drops. Because of this the pumping power is an appreciable fraction of the reactor output and in a gas cooled reactor providing electric power it can amount to 10% of the electrical power output. A high pressure drop in this context is of the order of 3–4 lb/in.^{2} so that even a fraction of a pound introduced by a venturi or orifice increases the total pressure drop of the circuit perceptibly. This must be reduced to a minimum and sensitive pressure measuring devices used. Alternatively an orifice plate can be inserted temporarily as a means of calibrating the pressure drop across some component in the circuit in terms of coolant flow. The flow in the very large gas ducts necessary in order that the circuit pressure losses be kept down may be not uniform across the section and the accuracy obtained even with an orifice plate is questionable, and for the most accurate calibration a traverse across a section with Pitot tubes should be made.

For reasons of safety or because of the difficulty in passing large volumes of coolant through one cooling circuit, multiple circuits are used. They have, of course, a common path through the reactor core. Unless the measuring signal can

be obtained in the common circuit, the flow in each circuit has to be measured and the total flow obtained by summation. Usually the coolant pressure and temperatures are held within close limits so that correction for temperature and pressure can be made simply. Care has to be taken so that in the event of mechanical damage or failure to the detecting system, material which would contaminate the coolant or reactor cannot get into the coolant stream. Because of this, pneumatically operated devices are to be preferred to those employing hydraulics or mercury.

3.3 HEAT OUTPUT

A measurement of the total heat output is the only absolute method of determining the reactor power. In power reactors most of the heat is lost to the coolant but as mentioned above some is lost by thermal radiation, conduction and neutron and gamma radiation to the containing vessel and biological shield. These structures are cooled and so it is possible to measure the heat given up by them to this coolant.

A typical heat balance for a large reactor would be:

To core coolant directly	99·5%
Heat loss from vessel other than due to neutron heating	0·03%
Neutron and gamma radiation	0·47%
Total power output	100·0%

Because of the large masses of material in the shielding structure it takes a long time to reach steady thermal conditions so that an accurate heat balance can only be made with the reactor running at a steady power for a long period. If the temperatures in the reactor are held sensibly constant, irrespective of power level, the heat loss from the containing vessel, apart from that due to neutron heating, is constant. The neutron heating is approximately proportional to output so that by measuring the heat passed into the reactor coolant, correcting proportionately for the loss due to nuclear energy and adding a fixed correcting factor for the direct heat loss, the total power can be estimated very closely. For operational work, however, these small corrections can be neglected.

The heat taken up by the coolant is given by

$$H = MC_p(T_2 - T_1) \qquad (3)$$

The mass flow can be determined by methods discussed above and the temperature rise by opposing thermocouple or resistance thermometers. The specific heat of most coolants vary with temperature and if the inlet and outlet temperatures are not held constant a correction should be made for the specific heat variation. Satisfactory instruments have been developed to do this computation.[8] They employ standard potentiometer recorders recording flow and temperature difference fitted with extra potentiometer slide wires. By using the output of one potentiometer to supply another multiplication is effected. A single self balancing resistance bridge has also been used to perform the multiplication. The overall response of these instruments is determined mainly by that of the detectors which need not be more than a few seconds.

3.4 OTHER PHYSICAL MEASUREMENTS

Apart from the measurement of temperature, coolant flow and power, other instruments are required to monitor the pressure of the coolant, leakage of it and leakage of material into it. Because the consequence of leakage into or from the cooling circuit can be serious these instruments must operate continuously and give early warning of any change in conditions and in some cases shut down the reactor.

Leakage of coolant can be detected by change of pressure but additional protection can also be afforded by fitting instruments at most likely sources of leakage such as seals of sliding or rotating shafts in the pressure circuit. Leakage into the coolant can cause corrosion, rise in activity of the coolant or, in the case of liquid metal coolants, fouling and blockage of the cooling circuit. Contamination of the coolant from inside the reactor is discussed in Section 5. The most likely source of leakage from outside is from a secondary cooling circuit and a dectector sensitive to the secondary cooling medium is generally fitted in the primary circuit. The secondary coolant is most commonly water and for which fortunately many sensitive detectors are available.

4.0 SAFETY

It is of vital importance that at all times a reactor is protected against damage due to maloperation or a failure of a component of the reactor or associated equipment. With low energy reactors the primary concern is to prevent the reactor getting out of control and the power rising to a dangerous level. For power reactors it is equally essential to ensure an adequate supply of coolant. Whether the fault results in a power surge or insufficient flow of coolant, overheating occurs. In other words there is a maximum safe ratio of power generated to coolant flow above which overheating will result. Instruments must be provided to indicate faults, operate alarms, and if need be trip circuits. Some of the safety and alarm signals can be conveniently provided by the normal measuring instruments and recorders but where these are not suitable, special safety instruments and detectors have to be provided. Care must be taken in both the design of the instruments and the way in which they are used that, should they fail, the reactor is not jeopardized thereby. On the other hand, accidental shut down of a reactor due to instrument failure can be expensive: the loss in revenue from a reactor in a nuclear power station would be several hundred pounds an hour and the loss with a large experimental reactor can be of the same order based on capital charges alone, but to this must be added the waste of technical labour and delay in obtaining results of experimental work. Any precautions taken against accidental shut down should not, however, impair the safety system. When an instrument which is complex, and therefore more prone to failure, is needed in a safety circuit, two or more units are normally fitted for the same duty, so arranged that the reactor is tripped only when at least two instruments indicate fault conditions.

It is also important to be able to test the safety circuits. With the above coincident arrangement a complex safety circuit can be tested completely without tripping the reactor. Single circuits can only be tested section by section and for this reason some instruments are duplicated even though this is not warranted on the score of reliability. Another desirable feature of safety arrangements is detecting a fault by more than one type of instrument or circuit. For example, although excess power would normally be detected by a flux measuring instrument, it could also be shown up by a suitably placed thermocouple. The response of the latter would, however, be slower. Again, a relay could detect failure of an electrical supply to a coolant pump and at the same time an

alarm could be initiated by the reduction in flow monitored by a flow meter.

In addition to the reduction of the chance of inadvertent shut down, guaranteeing the instrument supplies contributes to the safety of the plant in that under fault conditions the operator is provided with full information and so has a better chance of mitigating the effects of the fault. As a further safeguard it is usual to provide the operator with indications of power and vital temperatures by instruments which do not require external supplies. Unfortunately these instruments are rarely suitable for direct operation of trip circuits.

5.0 DETECTION OF FAULTS IN FUEL CONTAINERS

Most power reactors constructed so far use solid uranium fuel from which the heat is removed by forced cooling. Uranium reacts chemically at elevated temperatures with nearly all coolants and to prevent this it must be sealed in containers of material compatible with the coolant and the uranium. In addition, fission products which are radioactive escape from the surface of the uranium and if released would contaminate the coolant and cooling system. Thus even if the coolant and uranium do not react, it is desirable that it be contained.

In experimental reactors running in air at room temperature and generating only a few watts, containers for the fuel are not necessary because the reaction between uranium and air at these temperatures is very slight and the quantity of fission products released is negligible. If the release of material, either due to chemical reaction or by fission, is to be kept to a minimum it is essential that a fault in any container be detected as early as possible. This calls for a detector system with as high a sensitivity as possible. It is not sufficient to know that a fault has occurred: the fuel containers are generally in channels along the length of the core and it is necessary to be able to identify at least the channel in which the fault has occurred if not the actual container. In power reactors in which there may be thousands of channels, whatever system is used, the equipment is likely to be mechanically complex and an appreciable fraction of the whole reactor instrumentation.

Two main approaches have been made to the problem. One is to make use of physical changes and the other to measure the activity released through the fault, small though it may be. The only satisfactory physical method has been to measure the loss in pressure of gas in the container due to its escape through a fault. Changes in flow of coolant due to deformation of the fuel and/or containers have been suggested but unless the cooling channels are very small this method is not very practicable or sensitive and in any case presupposes the nature of the deformation. There are three possible methods of implementing the second approach: detection of neutrons emitted from fission products, gaseous fission products or gamma activity of material released into the coolant.

In spite of the high energy imparted to them in the fission process, the fission products can only travel of the order of 5×10^{-3} mm in the uranium before coming to rest. And so only products of fissions occurring near the surface of the fuel have a chance of escaping. In any fission the tendency will be for one fragment to be driven outwards and the other inwards so that at best only half will escape. Assuming that one fragment of each fission in the outer skin 5×10^{-3} mm deep escape from uranium operating at 1 W/g, approximately 3×10^{8} fragments would be escaping from each square centimetre of surface per second but because some of the fragments travel obliquely and do not reach the surface, the rate is not quite as high as this.

5.1 DETECTION OF DELAYED NEUTRONS

In the fission process 99·3% of the neutrons are released directly. The remaining 0·7% are produced later by the decay of unstable fission products. There are five fission products produced in significant amounts which emit neutrons but as the half-lives of two of them are short, 0·43 and 1·52 s respectively, they contribute little by the time they arrive at the detector if it is more than 1–2 s transport time away. The others, ^{45}Br, ^{137}I and ^{87}Br, produce about 0·5% of the neutrons. The number of atoms of these isotope escaping into the coolant per second from 1 cm^2 of fuel running at 1 W/g are given in Table 2. The initial rate of emission of neutrons from them is also shown.

Table 2. *Delayed neutron emitters released into coolant from 1 cm^2 of uranium rated at 1 W/g*

Isotope	*Half-life* (s)	*Fraction of total neutron* (%)	*Atoms in coolant/s*	*Neutrons emitted/s*
^{89}Br	4·51	0·213	$6·6 \times 10^{5}$	100×10^{3}
^{137}I	22	0·166	$5·15 \times 10^{5}$	$16·2 \times 10^{3}$
^{87}Br	55·6	0·025	$0·78 \times 10^{5}$	$0·97 \times 10^{3}$

The coolant issuing from the channel or a sample of it could be passed through a neutron detector. This detector would have to be placed some distance from the reactor behind adequate shielding so that it is not affected by direct radiation from it. During the time the coolant will take to get to the detector, the rate of neutron emission will decay and thus the transport time should be kept as short as possible. The neutrons emitted are fast neutrons to which existing detecting chambers are not very sensitive and as the fission products other than iodine and bromium in the coolant stream emit gamma rays to which the fast neutron detectors are similarly sensitive, to obtain a signal due to neutrons alone some means of either removing the gamma emitters, or better, isolating the neutron emitters, must be employed. Until a satisfactory solution to this problem has been devised the delayed neutron method of detecting faulty containers will remain insensitive. Assuming this is achieved by physical or chemical means, the signals obtained are still likely to be quite small. During manufacture it is almost impossible to keep the outer surface of the fuel containers free from particles of uranium and in the rector these particles emit fission products into the coolant, the delayed neutrons from which will be detected. A background signal due to these neutrons, stray radiation from the core and cosmic effects will be present, the amplitude of which will set a lower limit on the area of exposed uranium which can be detected. For example, using the conditions as in Table 2, if the neutrons emitted from 5% of the coolant flow from a channel reached the detector in 10 s the neutron emission rate would be about 1000 n/s. The signal from the detector would be about 20 c.p.s and correspond to 1 cm^2 of exposed uranium. Assuming that 1 c.p.s. is significant, an area equivalent to 5 mm^2 could be detected.

A further interference signal is produced if oxygen is present in the coolant. The ^{17}O isotope of oxygen under irradiation produces the ^{17}N nitrogen isotope which decays with a half-life of 4·1 s to ^{16}O by the emission of a neutron. While the absorption cross-section of the ^{17}O is small, in the volume of coolant passing through the channel a significant

number of neutrons are produced. With a half-life of 4·1 s the decay of the ^{17}N is fairly rapid and the neutrons from this source may become insignificant after 10–20 s or so. By then, of course, the ^{89}Br will have decayed such that the detection is dependent on neutrons from the ^{137}I and ^{87}Br only.

5.2 DETECTION OF GASEOUS FISSION PRODUCTS

Xenon and krypton are formed directly in fission or as daughter products of other fission products. They decay to caesium and rubidium respectively which decay further by beta emission. Detection of this beta emission can be used as a measure of the xenon and krypton concentration in the coolant and hence an estimate made of the area of uranium exposed in a fault. Of over 30 isotopes of xenon and krypton formed in fission only 11 have appreciable yields and half-lives such that they are likely to contribute to the signal of a detector. Some of these isotopes are also produced by the decay of other fission products and this enhances their contributions. The half-lives of these isotopes together with the half-lives of the daughter products are shown in Table 3.

Table 3. *Gaseous fission products released into coolant from 1 cm² of uranium rated at 1 W/g*

Isotope	*Half-life* (s)	*Atoms released into coolant* × 10^5	*Daughter product*	*Half-life* (s)
^{88}Kr	11 000	1200	^{88}Rb	1080
^{89}Kr	150	6000	^{89}Rb	900
^{90}Kr	30	6000	^{90}Rb	162
^{91}Kr	9·3	4800	^{91}Rb	100
^{92}Kr	2·3	3000	^{92}Rb	$\simeq 1$
^{93}Kr	2·2	1500	^{93}Rb	< 1?
^{94}Kr	1·4	350	^{94}Rb	$\simeq 1$
^{138}Xe	1000	4000	^{138}Cs	1980
^{139}Xe	41	4500	^{139}Cs	480
^{140}Xe	16	4500	^{140}Cs	50
^{141}Xe	1·7	2400	^{141}Cs	< 1?

Apart from the effect due to the production of xenon and krypton as daughter products, the rate of production of the rubidium and caesium and the subsequent amount of beta emission is a maximum at the time the xenon and krypton are released from the uranium, consequently as when detecting delayed neutrons, the shorter the transit time to the detector the larger the signal. Practical difficulties limit this to at least 5 s. The decay chains which are likely to contribute most to the beta emission after this time are those in which the half-life of the decay to the daughter product is shorter or of the same order as, and the half-life of the daughter appreciably longer than, the 5 s. The yields of the various isotopes also, of course, affect their concentration. From Table 3 it can be seen that the most useful contributors are likely to be ^{91}Kr and ^{140}Xe.

In a gas cooled reactor to reduce interference from other fission products, or other active particles in the coolant, filters have to be inserted before the detector. This, however, reduces the signal due to the xenon and krypton which have already decayed in transit from the reactor and which have attached themselves to solid particles. The filtered coolant could be then monitored for beta emission but the sensitivity is considerably increased if the gas is fed through a chamber containing a wire held at a high potential. The particles are precipitated on to a wire which is passed under a β-ray detector. The method can be adapted for use with liquid cooled reactors if a stream of gas is passed through the liquid to "leach out" the gaseous fission products. By this method faults as small as 1 mm² have been detected.

5.3 DETECTION OF GAMMA-RAY EMISSION

The gamma-ray emission of fission products can be used to detect exposed uranium provided the background gamma signal from other sources is not too high. Small particles of dust carried in the coolant stream can become very active and the coolant itself may produce gamma rays. A physical filter can remove the solid particles and some of the fission products which are trapped on the filter not by direct action but by their attachment to the dust particles. Monitoring such a filter can indicate a rise in activity in the coolant, particularly if some uranium oxide or other compound formed by chemical action with the coolant becomes detached and is deposited on the filter.

6.0 ESSENTIAL INSTRUMENTATION OF A POWER REACTOR

As an illustration of the ways in which the various instruments described in previous sections are employed, the essential instrumentation of a power reactor is shown in Fig. 8. The reactor assumed is a graphite moderated unit rated at 200 MW.

The site for the thermal column has often to be a compromise between nuclear and structural requirements with the result that the fluxes in the detecting chambers are lower than the optimum. The ideal would be to have a flux of 10^{11} n cm^{-2} s^{-1} at the detectors with the reactor at full power of 200 MW. It has been assumed for the example that the maximum flux available is one order less than this and that neutron sources in the core raise the shut down power to 2 W. The shut down neutron flux at the detectors is therefore 100 n cm^{-2} s^{-1} in which an RC1 type ion chamber could provide a current of $1{\cdot}5 \times 10^{-11}$ A. This is comfortably within the lower limit of 10^{-12} A that can conveniently be amplified but even when employing gamma shielding the current due to interference from gamma-ray activity external and induced in the chamber is likely to be of the same order as that due to neutrons even some time after shut down. Counter equipment is therefore provided for low power measurements. Two logarithmic channels cover the measuring range with an overlap of four decades. Coverage of the upper three decades can be achieved by the linear power channel with measuring resistors not exceeding 5 MΩ. The power deviation is monitored from 1 MW to full power. The three safety circuits covering from 10 MW upwards operate the trip circuits through coincidence relays. The logarithmic and counter circuits all provide period trips.

The temperature measuring facilities are similar to those described in Section 3. Over 100 thermocouples measure the fuel and coolant temperatures in the core and twice this number are used for the core and containing vessel. Some 500 thermocouples are embedded in the biological shielding although only a small proportion would normally be monitored.

Each of the multiple cooling circuits are monitored for flow, temperature rise and coolant purity. The total flow and heat output are also measured.

The amount of equipment required to detect faulty fuel containers is determined by the frequency that it is deemed necessary to check each fuel channel and by the time required to make the measurement. The ten units shown are for a core of 2000 channels checked every hour by detectors with approximately 15 s resolution time.

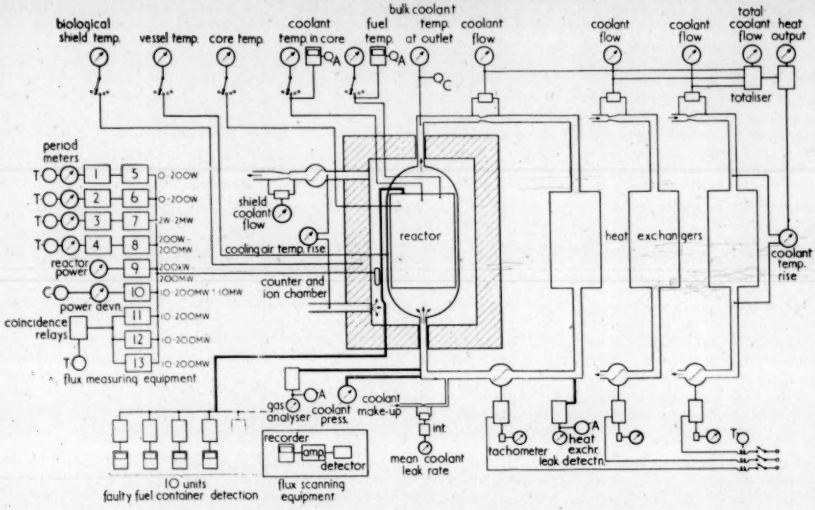

Fig. 8. Schematic diagram of essential instrumentation of a graphite moderated gas-cooled power reactor
A, alarm; *C*, control; *T*, trip.

7.0 FUTURE DEVELOPMENTS

As instrumentation is ancillary to the reactor it is to be expected that its development will follow that of the reactor. It is suggested that two major features of future reactors designed for power production and experimental purposes will be greater simplicity of operation and inherent safety. The contribution that instrumentation can make to these will be partly born of operating experience with existing reactors. It will no doubt be found possible to dispense with some instruments and detectors and to reduce the numbers of others. Improvements in instrument design will also make their contribution. This is most likely in the field of flux measurement where better ion chambers and amplifiers will improve the range of individual measuring channels and extend the effective measuring range into that covered adequately at present only by pulse counting equipment. Work is now going on in this field and it is not too much to anticipate that the amount of flux measuring equipment necessary will be halved.

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APPENDIX

List of symbols

I = diode current
I_0 = diode current at zero voltage
e = electronic charge
v = diode voltage
k = Boltzmanns constant
t = absolute temperature of cathode
N = atoms/cm^3
σ_a = absorption cross-section in cm^2
ϕ = neutron flux in n cm^{-2} s^{-1}
λ = decay constant per hour
T = irradiation time hours
H = heat taken up by coolant
M = Mass flow of coolant
C_p = specific heat of coolant at constant pressure
T_1 = coolant temperature at reactor inlet
T_2 = coolant temperature at reactor outlet

DISCUSSION ON SESSION 6

Mr. C. C. Horton: The design of mobile reactors and competitive nuclear power plants had led to a considerable design effort being devoted to the design of reactor shields in the last few years. As a result, it may be said that the behaviour of γ-radiation in penetrating a shield is well understood, and the behaviour of neutrons qualitatively understood.

As Dr. Laurence points out, in neutron and γ-ray shielding, the penetration characteristics are determined by the highest energy photons or neutrons present; in the neutron shielding case the situation is further complicated by forward scattering effects at high energy in heavy nuclei. This leads to an apparent increase in the mean free path in the shield, a prediction confirmed by experiment. Calculation of shields with large hydrogen concentrations can be effected by the "removal cross-section" technique introduced by Welton. This utilizes the fact that in a thick shield any collision which removes a neutron from its outward path is equivalent to an absorption due to the subsequent high energy loss in collision with the hydrogen present. This method breaks down if the hydrogen content is very low.

In present-day high flux reactors, the heating and damage effects in the shield are of importance, the discussion given by Dr. Laurence quotes some useful criteria here. Other related effects may also be of importance; for example, radiation decomposition of a water shield may lead to pockets of gas in the shield which reduce the effectiveness of the shield.

There are several points worth mentioning in general shield design which are sometimes overlooked.

(*a*) The dose-rate allowable at the outside of a shield is determined by the necessity to protect the operating staff; much weight and cost can be saved by control of personnel movement.

(*b*) The design of a neutron shield should make adequate allowance for the presence of capture γ-radiation. For example, it can be shown by use of equation (8) of Dr. Laurence's paper that the capture γ-radiation from a typical water neutron shield may contribute a dose several orders of magnitude worse than the neutron radiation.

(*c*) Finally, when designing an economical shield, it should be borne in mind that the overall cost is influenced by many factors. For example, the cost of the reactor facility is increased as the shield thickness is increased owing to the extra height and width of the building, building foundations, longer remote handling tools, and so on.

Dr. Denis Taylor: At this meeting the speakers have concerned themselves very largely with power reactors. Because of this a great deal of emphasis, and rightly so, has been placed on the measurement of temperature, coolant flow, heat output, etc., but proper consideration is given in the papers to the more novel nuclear measurements, e.g. neutron flux, β- and γ-activities, etc.

There is naturally some interest in reducing the instrumentation needed and, from his final remarks, Mr. Macrae appears to be looking forward to simplification of the nuclear measuring equipment, particularly by improvement in the performance of the detectors.

It is perhaps appropriate to mention that another author in a recent paper,[1] mentioned that the current need in reactor instrumentation is in detector development rather than in the transmission and recording of the data. This also appears to be the views of the present authors. However, the paper I have just mentioned then goes on to say that the most troublesome detectors thus far have been the thermocouples for fuel plate temperature measurements. This difficulty is mentioned in Mr. Macrae's paper, but I wonder whether he would agree that it represents the most serious instrument problem at the present time. The difficulty appears to be primarily a mechanical one resulting from the rapid differential expansion between the fuel plate and the thermocouple wires. It also has to be remembered that facilities have to be provided for loading and unloading the fuel elements which must complicate the temperature measurements.

I have wondered in listening to the authors this afternoon if the uninitiated might have the impression that the circuit arrangements described by the authors have now become standard. If this impression has been conveyed I would like to correct it. Certainly the circuit arrangements described have been most carefully tested, but the subject is still very new, and new and improved systems will continue to be developed. It is reasonable to point out perhaps that the use of a pentode valve to give the logarithmic response as required for the period meter was used experimentally for the first time (although in connection with a different non-reactor application) in 1951, and a patent was not applied for until 1953. We have used period meters of this type experimentally in BEPO for some time, but the first full-scale reactors to have these period meters installed as part of the main instrumentation will be DIDO and the Calder Hall power station. The first is not yet commissioned and the second has so far only worked at low power. Naturally the equipment has been carefully tested in the laboratory, but we are still lacking lengthy operational experience. This also applies to some of the other circuits.

Dealing now with Dr. Salmon's paper, I was interested to note that he has used the orthodox approach of writing down his kinetic equations in terms of the neutron density. In a recent paper[2] Cox and Walker express the same equations in terms of the neutron population. Some of us feel that there are advantages in this method, but I would be interested in Dr. Salmon's opinion.

In the section on "delayed neutrons" Dr. Salmon mentioned that at small reactivities all reactors behave similarly from the control point of view, but that after the reactors become prompt critical their behaviour becomes very different. This fact that fast and thermal reactors are indistinguishable from the operator's viewpoint (each reactor having the same response to similar changes in control movement for small reactivities) is brought out well in Fig. 1, which shows the connection between the reactivity ρ and the reactor period T for various Harwell reactors.

Dr. Salmon's paper contains the results of a good deal of computation work, and I had hoped therefore that he would have dealt with the important problem of rates of reactivity addition to achieve a safe, but reasonably fast start-up, but his paper gives no quantitative data. His comments on this matter would be welcome.

Dealing now with Mr. Macrae's paper, I think that he has been a little unfair on the BF_3 proportional counter. The individual pulses due to single γ-ray events are of course much smaller (of the order of 1/50th) than the neutron-pulses, but if there are a sufficient number of these pulses occurring within the response time of the equipment the resulting pile-up of gamma pulses may be indistinguishable from neutron pulses. The gamma dose-rate at which it is possible to use a BF_3 counter will depend on the counter size and filling and on the differentiating and integrating time constants of the pulse amplifier. With short amplifier time constants (e.g. 0·2 μs) and a 6 × 1 in. counter it is possible to measure

neutron flux in the presence of γ-fields of a few hundred roentgens per hour. I agree that fission counters of similar size may be used at much higher gamma-radiation levels because of the increased size of neutron-pulses (from fission particles) relative to individual gamma-pulses. It is also appropriate to

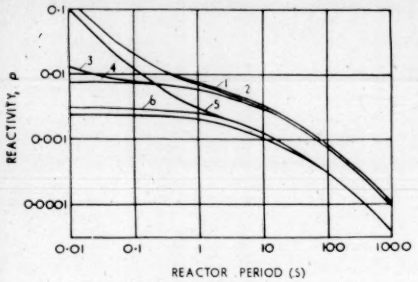

Fig. 1. Relationship between reactivity ρ and reactor period T for typical systems (from Ref. 2)

Curve 1. DIDO. Curve 5. Theoretical (^{239}Pu thermal).
Curve 2. BEPO. Curve 6. ZEPHYR.
Curve 3. LIDO. Curve 7. Theoretical (^{239}Pu fast).
Curve 4. ZEUS.

mention that we have not used fission counters for the Calder Hall installation; BF_3 counters have been used because of the higher sensitivity available in a given size counter.

Mr. Macrae emphasizes the need for measuring equipment which is independent of external supplies and can therefore provide power indication at all times (e.g. even in the event of mains supply failure) and he is very enthusiastic about the galvanometer system for power level indication for this reason. However, many of the alternative methods using electronic amplifiers are, or can be very economical in power supplies, and can if required be made independent of external supplies. Systems using transistors in place of thermionic valves are also possible. It would be of interest to know whether Mr. Macrae would welcome these alternative systems.

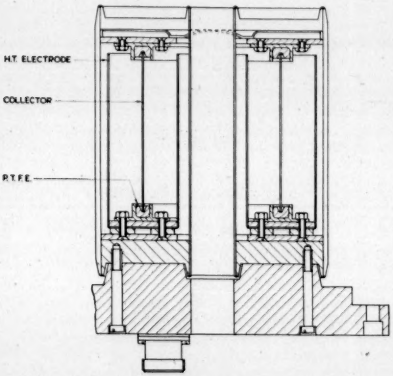

Fig. 2. Ionization chamber for wire activitation measurements

Finally, I would like to give some information about the special purpose radiation detectors used for flux scanning measurements. Fig. 2 shows the type of ionization chamber used for the measurement of wire activation. This method was mentioned by Mr. Macrae. The wire passes through a 1 in. hole down the centre of the chamber. This detector is suitable for measuring γ-activities such as would result using a tungsten wire. In some cases it has proved necessary to measure β-activities. The chamber is then somewhat smaller and the inner wall needs to be quite thin ($\sim$0·002 in.) and the hole is only 0·25 in. diameter.

Fig. 3 shows the 0·25 in. diameter fission chambers of the type designed for flux measurements along the length of the

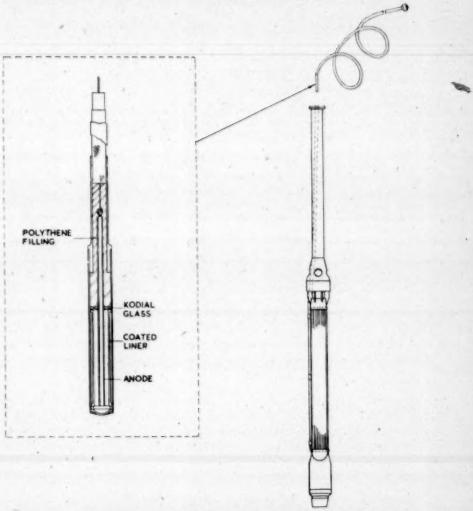

Fig. 3. Fission counter for flux measurements along the length of fuel element

DIDO type fuel element. It will be noted that these counters have to be threaded down a long tube into the fuel element assembly.

Mr. D. Little and **Mr. M. A. Schultz** (*presented in summary by Mr. Schultz*): A study of the non-linear kinetics of heterogeneous reactors can lead to basic design criteria which are useful in designing stability into a given reactor. The design of most reactors is usually based upon other physical parameters such as reactivities, heat transfer surfaces, metallurgical conditions, and operating lifetimes. The question of reactor stability is often deferred on the design schedule until these other parameters have so closely bounded the design that only incidental changes are possible. It is fortunate that many reactors are naturally stable over wide ranges of design conditions.

Non-linear kinetics have been examined in two ways in the past. The first method is to modify the basic reactor kinetic equations to include the effects of temperature, pressure, or poisoning. Solutions of these equations generally can be obtained with some approximation being needed. The second method consists of obtaining the transfer function of a simple

reactor and to modify the characteristics of this reactor by feedback networks. The first method has been used successfully by Weinberg and Ergen[3] for homogeneous reactors and by Lipkin[4] and others for heterogeneous reactors. The second method is an engineering one which is capable of giving considerable information over a wide range of variables and will be used in this contribution with suitable approximations to examine the stability of heterogeneous reactors.

The basic reactor kinetic equations can be of the form[5]

$$\frac{dn}{dt} = \frac{\delta k - \beta}{l^*} n + \sum_{i=1}^{6} \lambda i C i \qquad (1)$$

$$\frac{dC_i}{dt} = \frac{\beta_i}{l^*} n + \lambda_i C_i \qquad (2)$$

where n = the neutron level, directly related to the reactor power level
δk = the reactivity
l^* = the mean neutron life
β = the total fraction of the delayed neutrons
β_i = the fraction of the delayed neutrons in the ith group of delayed neutrons
C_i = the concentration of the delayed neutrons in the ith group.

From these equations, the transfer function representing the small signal sinusoidal response of the reactor has been derived[6] in Laplace notation as

$$\frac{\delta n(s)}{\delta k(s)} = \frac{n_0 \prod_{i=1}^{6} (s + \lambda_i)}{l^* s \prod_{i=1}^{6} (s + r_i)} \qquad (3)$$

where $\delta n(s)$ = a small sinusoidal variation in neutron level about a steady state value n_0, s = the Laplace operator $= j\omega$ for the linear operation indicated and r_1 = a constant depending upon β and l^*. Fig. 4 shows graphically this transfer function in amplitude and phase for a reactor having an $l^* = 10^{-4}$ s. An approximate transfer function of the form

$$\frac{\delta n(s)}{\delta k(s)} = \frac{n_0(s + \bar{\lambda})}{sl^*(s + \bar{r})} \qquad (4)$$

where $\bar{\lambda}$ and $\bar{r}$ are average values of the delayed emitter constants will be used later for analysis purposes. The validity of this approximation is indicated by the dotted curves of Fig. 4 when $\bar{\lambda} = 0{\cdot}125\ s^{-1}$ and $\bar{r} = 50\ s^{-1}$. Equations (3) or (4) or Fig. 4 completely described the linear kinetics of a given reactor from a servo point of view. To examine the non-linear performance of the reactor, we must hypothesize feedback loops around this basic reactor transfer function. This analysis will ignore the effects of poisons and will examine only the effects of changing temperature. Poisons present a problem on a different stability time scale than does temperature and consequently can be treated separately.

In a single region analysis of a heterogeneous reactor, changing temperature can cause several types of reactivity changes. A block diagram of the transfer function of a simple reactor and its thermal feedbacks might appear as given in Fig. 5. This figure represents a testing reactor such as the Westinghouse Testing Reactor containing a fuel

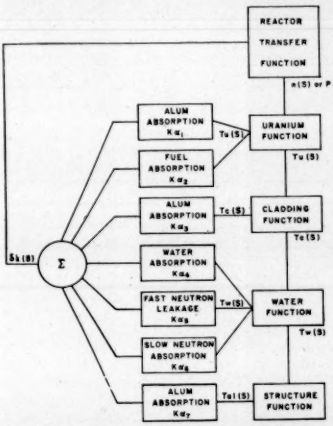

Fig. 5. Block diagram indicating temperature coefficient feedback paths (Westinghouse Testing Reactor)

element composed of a uranium–aluminium alloy clad with aluminium. The moderator and reflector are water, and the fuel elements are supported in a grid-like structure to make up the core. From Fig. 5, the output of the reactor is indicated as being neutron level or power level. An upward change in this neutron level causes heat to be generated in the uranium, and shortly thereafter the temperature of the uranium fuel alloy rises. The heat is then transferred to the cladding, and again after a short time delay the cladding temperature increases. Further transfers of heat are made to the water and then to the structure of the core. The fuel, the cladding, the water, and the structure all change their reactivity characteristics somewhat as the result of these temperature changes, and it is the sum of these reactivity changes as a function of temperature which defines a gross reactor temperature coefficient. For the reactor of Fig. 5, changes in the fuel temperature cause changes in the neutron absorption cross-sections of uranium and aluminium. Changes in the water temperature cause water cross-section changes and affect the fast neutron and slow neutron leakage. Finally, changes in the structure can cause changes in neutron absorption and dimensional changes in the reactor. To each one of these changes may be assigned a local temperature coefficient α, which may be positive or negative. The net

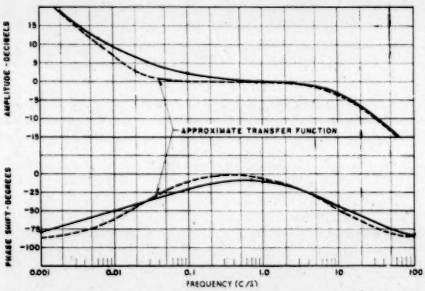

Fig. 4. Reactor transfer function, amplitude and phase, $l^* = 10^{-4}$ s

reactivity feedback effect is obtained by summing up all of the individual reactivity changes as a function of frequency. The block diagram of Fig. 5 giving the transfer function representation of these temperature feedbacks is quite complex but may be solved graphically or by an analogue computer providing the various temperature coefficients and time constants can be obtained.

A useful analytical solution can be made if the values of time constants involved in the heat transfer processes are such that the temperature feedback can be approximated by two simple paths, one representing the fuel and the other the water. This approximation is quite valid in some cases. For example, in Fig. 5 the time constant of the cladding might well be much shorter than that of the fuel alloy, and consequently the two transfer functions might be combined. Fig. 6 gives a simplified block diagram for this type of representation where all of the multiple paths of Fig. 5 have been resolved into two feedback paths.

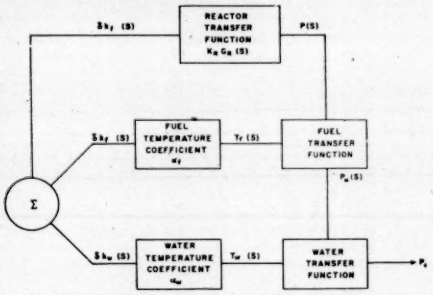

Fig. 6. Block diagram of two feedback loop system

The time dependent equations associated with these simple feedbacks represented in Fig. 6 are

$$P = \mu_f \frac{dT_f}{dt} + P_w \tag{5}$$

$$P_w = \mu_w \frac{dT_w}{dt} + P_e \tag{6}$$

$$P_w = \xi(T_f - T_w) \tag{7}$$

where T_f = fuel temperature
T_w = water temperature
$\mu_f = W_f C_f$ = the weight of the fuel times the specific heat of the fuel
$\mu_w = W_w C_w$ = the weight of the moderator times the specific heat of the moderator
ξ = a heat transfer coefficient between fuel and water. ξ is generally a function of power level but for the small sinusoidal changes to be used here will be a constant
P_e = the power output taken from the water-moderator-coolant which will also be assumed to be constant at any convenient reactor output such as full power
P_w = the heat transferred from the fuel to the water.

These three equations give an elementary lumped picture of the heat transfer situation. And from Fig. 6, after the temperatures as a function of frequency have been derived, they are multiplied by local temperature coefficients to get the reactivity feedbacks.

By considering only small sinusoidal variations about a fixed power level, equations (5), (6) and (7) can be linearized and combined to give $T_f(s)$ and $T_w(s)$ in Laplace notation as

$$T_w(s) = \frac{\delta P(s)\xi}{s[\xi(\mu_f + \mu_w) + \mu_f \mu_w s]} \tag{8}$$

$$T_f(s) = \frac{\delta P(s)\,(\xi + \mu_w s)}{s[\xi(\mu_f + \mu_w) + \mu_f \mu_w s]} \tag{9}$$

The total feedback reactivity

$$\begin{aligned}\delta k_t(s) &= \delta k_f(s) + \delta k_w(s) \\ &= \alpha_f T_f(s) + \alpha_w T_w(s) \\ &= \frac{\delta P(s)[\xi(\alpha_w + \alpha_f) + \alpha_f \mu_w s]}{s[\xi(\mu_w + \mu_f) + \mu_f \mu_w s]}\end{aligned} \tag{10}$$

and the transfer function of the combined reactivity feedback becomes

$$K_F G_F(s) = \frac{\delta k_t(s)}{\delta P(s)} = \frac{\xi(\alpha_w + \alpha_f) + \alpha_f \mu_w s}{s[\xi(\mu_w + \mu_f) + \mu_f \mu_w s]} \tag{11}$$

This transfer function is of the form

$$K_F G_F(s) = A \frac{(1 + \tau_1 s)}{s(1 + \tau_2 s)} \tag{12}$$

where

$$A = \frac{\alpha_w + \alpha_f}{\mu_w + \mu_f}, \quad \tau_1 = \frac{\alpha_f \mu_w}{\xi(\alpha_f + \alpha_w)}, \quad \tau_2 = \frac{\mu_f \mu_w}{\xi(\mu_f + \mu_w)} \tag{13}$$

It will be noted that τ_2 is always positive, but A and τ_1 can be either positive or negative depending on the signs of the local temperature coefficients.

Fig. 7 now represents the simplified block diagram of the reactor and its overall temperature feedback. To examine the stability of the system represented by this closed loop, we can employ the familiar Nyquist plots of the open loop

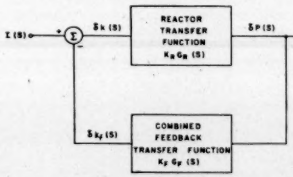

Fig. 7. Simplified block diagram of reactor and overall temperature feedback

transfer function. The open loop transfer function using the approximate reactor transfer function of equation (4) becomes

$$K_R G_R(s) K_F G_F(s) = C \frac{(1 + 1/\bar{\lambda}s)(1 + \tau_1 s)}{s^2(1 + 1/\bar{r}s)(1 + \tau_2 s)} \tag{14}$$

where

$$C = \frac{A\bar{\lambda}n_0}{\bar{r}l^*}$$

A number of interesting cases now must be examined to see over what range of temperature coefficients the system is stable. It will be recognized from the sign convention of the block diagram of Fig. 7 and the closed loop feedback transfer function equation

$$\frac{O(s)}{I(s)} = \frac{K_R G_R(s)}{1 + K_R G_R(s) K_F G_F(s)} \tag{15}$$

that the usual sign convention for α_f and α_w has to be reversed in using equation (14). That is, if α_f represents a negative temperature coefficient, it is inserted into equation (14) as a positive number and vice versa.

Various values and ranges of local temperature coefficients can now be inserted into equation (14). The stability situations and generalized forms of Nyquist plots which result from combinations of α_f and α_w are indicated in Fig. 8.

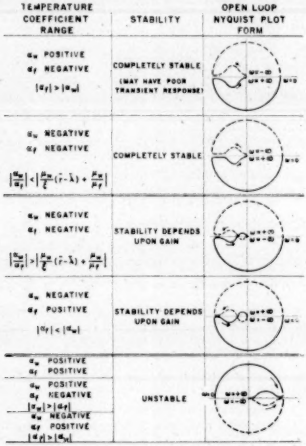

Fig. 8. Reactor stability for various ranges of local temperature coefficients

There are evidently ranges of coefficients over which the reactor loop can be either completely stable, stable depending upon gain, or completely unstable. The dependence of stability upon gain usually would be a function of the power level at which a specific reactor would operate.

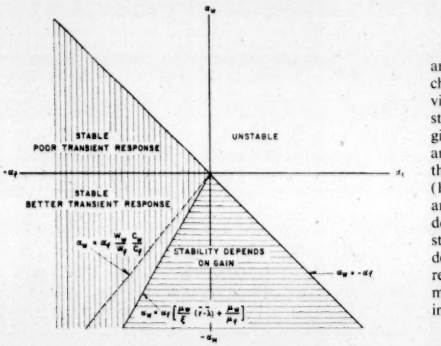

Fig. 9. Plot of α_f *vs.* α_w indicating stability regions associated with various parameter values

The stability situations thus obtained can be summarized by plotting α_w against α_f as indicated in Fig. 9. From an elementary physical point of view if both α_f and α_w are positive, that is, there is an overall gross positive temperature coefficient, the loop is obviously an unstable one. Or if the sum of a positive temperature coefficient and a negative temperature coefficient in either combination of α_f and α_w is positive, then the loop again is unstable. These situations are indicated by the area to the right of the 45° line on Fig. 9.

Intuitively also, it is evident that α_f is the dominant temperature coefficient. Feedback through the fuel occurs quicker than feedback through the water because of the series time delay in transferring the heat from fuel to water. Consequently, it is not surprising to find that if α_f is negative and greater in magnitude than a positive α_w, the loop will always be stable. On the other hand, the completely stable situation does not exist if α_w is negative and greater in magnitude than a positive α_f. In this case the loop may or may not be stable, depending upon the gain of the system as given by C in equation (14).

For the case where both α_f and α_w are negative, unconditional stability exists over a wide range of variables as long as

$$\frac{1}{\tau_1} - \frac{1}{\tau_2} < \bar{r} - \bar{\lambda} \tag{16}$$

Substituting in the values of τ_1 and τ_2

$$\frac{\xi}{\mu_w}\left(\frac{\alpha_f + \alpha_w}{\alpha_f} - \frac{\mu_f + \mu_w}{\mu_f}\right) < \bar{r} - \bar{\lambda} \tag{17}$$

or

$$\frac{\alpha_w}{\alpha_f} < \frac{\mu_w}{\xi}(\bar{r} - \bar{\lambda}) + \frac{\mu_w}{\mu_f} \tag{18}$$

This equation then sets the upper limit on the relative amounts of negative temperature coefficient feedback which may be used in the reactor without the possibility of sustained oscillations.

For some reactor designs, $\frac{\mu_w}{\xi}(\bar{r} - \bar{\lambda})$ is small compared with $\frac{\mu_w}{\mu_f}$. And in any event, a very safe simple condition for design purposes for absolute stability in loop performance when both local temperature coefficients are negative is that

$$\frac{\alpha_w}{\alpha_f} < \frac{\mu_w}{\mu_f} \tag{19}$$

It will be recognized that the higher the negative feedback around the loop, the stiffer will be the response to transient changes in reactivity. In fact, from an overshoot point of view, it might be desirable to operate in the conditionally stable range, as here the best performance may result at any given power level. However, from both the safety aspects and transient response, the best overall system will be obtained the closer the reactor design is to the condition of equation (18). The temperature coefficients associated with the fuel and the water are not easy to obtain and there may be some doubt as to their accuracy. Therefore, the approximate stipulation of equation (19) seems to indicate a simple, safe design objective. From this equation for these applicable reactors in which the fuel element and the water can be made to represent the only effective negative feedback items in the core

$$\frac{\alpha_w}{\alpha_f} < \frac{|\text{Water}|}{|\text{Metal}|}_{\text{weight}} \frac{C_w}{C_f} \tag{20}$$

represents a safe design objective. From equation (20), this

design objective from a stability point of view can be achieved by setting up a proper water to metal ratio in the reactor. α_w and α_f are, of course, functions of the metal to water ratio, and the design criteria of equation (20) is obtained by a number of trial solutions. This stability design objective should impose few limitations on the reactor designer and will insure not having to worry about stability after other conditions have set the parameters of the core.

The method of analysis presented above for a simple reactor can be extended to handle more complex situations. These solutions can most easily be handled by computing machine techniques.

Mr. R. D. Harrison: Is any trouble encountered in the use of thermocouples owing to alteration of characteristics by radiation damage?

Dr. R. J. Wakelin: In any possible aircraft applications of reactors the total weight of the shielding must be an important factor. Could Dr. Laurence tell us whether any work specifically aimed at reducing this weight has been carried out?

Mr. D. G. Andrews: I agree with Dr. Laurence that in shieldings, semi-empirical methods are more likely to succeed than excessively theoretical work. If one takes into account the density variations in concrete and the leakage through gaps, factors of two or more may well be the rule. It is quite possible, however, to deal with some slots and holes by putting scatterer in the right places. For instance, in the design of a control rod gland, theoretical calculation of the size of the annular clearance to give a safe dose rate at the surface might well lead to an unacceptably small figure.

The practical approach would then be to increase the clearance to give the necessary running fit, and to add rings of scatterer at suitable positions between the gland and the core. This would reduce the fast neutron hazard, which usually dominates these cases.

In general, there is a need for more data on neutron and gamma spectra at entry to, inside, and leaving existing shields. Nevertheless, calculation work will always have its place not only in establishing the general magnitude of the significant radiation components, but also in checking that the reflected components are indeed negligible. Dr. Laurence has, however, hinted that the best way to solve a shieldings problem is to build it.

Dr. Salmon is to be congratulated on giving attention to other methods of control than the insertion of absorber into the core. Metallurgists and engineers who have put much thought into saving neutrons on cans, coolant and structures, cannot be other than unhappy in the knowledge that over a proportion of the life of a reactor, large numbers of neutrons are being uselessly absorbed in conventional control rods. Further work by physicists on alternative methods may help to make the absorbing control rod obsolete as soon as possible.

AUTHORS' REPLIES

Dr. A. J. Salmon: I feel, with Dr. Taylor, that eventually the neutron population may become the "orthodox" variable in the neutron kinetic equations. However, in a simple exposition of reactor control there is little to be gained by using population, as distinct from density, and perhaps there is something to be lost as its use prompts questions which cannot be fully answered in a simple discussion. With regard to the question about the maximum possible rate of reactivity addition a possible criterion for this rate can be stated. The maximum rate is that which if continuously added to the maximum investment of the control units being considered, will still not produce a dangerous condition of the reactor. Unfortunately, this problem is not always susceptible to theoretical treatment, even using computors as an aid to calculation, and experimental data is not available. In such cases, a conservative rate of reactivity addition will have to be used or stringent safety precautions will have to be made. The reactor transient experiments on the BORAX and SPERT reactors in the U.S.A. are providing very useful data in this respect.(7)

Mr. Little and Mr. Schultz are to be thanked for their comments on temperature effects, thermal delays and the stability of reactors. We are at the beginning of a period during which many papers will be produced on this topic of reactor stability and transient effects. The experiments on BORAX and SPERT, mentioned above, are part of this important trend.

With regard to current requirements in reactor instrumentation it seems to me that two important developments are needed in the United Kingdom. The first is a simple and sensitive fission product detector for water-cooled reactor systems. The second is a γ-ray compensated neutron detector, this is especially required in systems where space is limited.

Mr. W. Macrae: I am indebted to Dr. Taylor for enlarging on a number of points on measuring technique. He asked whether difficulties with thermocouples for fuel element temperature measurement is the most serious instrument problem at the present time. It is not possible to be specific on this point, but it is a major one in power reactors. As present fuel containers have to be run near their ultimate metallurgic limit of temperature, accurate measurement of their temperature is essential. The main difficulty as I see it, is bringing out the leads to the measuring points, not the attachment of the junctions to the containers.

As far as I am aware no ageing effects of thermocouples under radiation have been reported.

I agree with Dr. Taylor that the overall neutron to gamma ratio with boron trifluoride proportional counters can be improved with suitable amplifiers. Their notable feature is, however, their considerably higher sensitivity than fission chambers enabling much lower power levels to be measured accurately.

The power consumption of present flux measuring equipment for a power reactor is such that battery supply is not practicable. The approach has been to guarantee the supply to these instruments as far as possible. This is not entirely satisfactory as all channels have a common link in this supply. If transistor amplifiers with low current consumption were developed they would indeed be a welcome alternative. In fact, a beginning has already been made in the use of transistors in monitor equipment for ionization chamber battery supplies.

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SESSION 7. EVENING LECTURES

Chairman: B. F. J. SCHONLAND, C.B.E., Sc.D., F.R.S.

The role of chemistry in a nuclear energy project

By R. SPENCE, C.B., D.Sc., F.R.I.C., Atomic Energy Research Establishment, Harwell, Berkshire

The development of atomic energy has been unique from a chemical point of view, in that a whole series of new elements, the actinide series, has been produced and some of these, such as neptunium, plutonium and americium, play a major role in the technology. Furthermore, many elements which were previously little known and of academic interest only, have become of technical importance in the project, as for example, ruthenium, zirconium and protactinium.

Atomic energy can be divided into the following broad fields, from the chemical point of view, viz., raw materials; production of fuel elements and other special reactor materials; reactor chemistry; chemical processing of irradiated materials; disposal and utilization of radioactive waste materials.

Although the provision of materials and the processing of irradiated fuel elements are of great importance, the nuclear reactor itself has always been the centre of interest. The earlier forms of reactor did not in themselves present any very serious chemical problems. Newer forms of reactor, however, operate at high temperatures and frequently bear more resemblance to chemical or metallurgical plants than they do to mechanical apparatus.

Chemists have been involved in the development of radioactivity and nuclear physics since the discovery of radium by Madame Curie. It was necessary at that time to separate radium and to study its chemical properties in order to arrive at a better understanding of the process of radioactive decay. Subsequently other members of the natural radioactive series were isolated in order to provide pure specimens for nuclear physical studies and in order to learn more about the properties of hitherto inaccessible elements. It was also found that radioactivity had a special significance to chemistry for other reasons. The extraordinary sensitivity with which radiation can be detected together with the possibility of mixing radioactive isotopes with inactive isotopes of the same element led to new studies of the behaviour of elements present in trace concentration and to the now widely applied method of radioactive tracers. All these fields, involving the study or use of radioactive substances, have tended to be grouped under the general heading of Radiochemistry. Another important field, quite distinct from radiochemistry, namely the study of the chemical effects of nuclear radiation, known as Radiation Chemistry, was also recognized at quite an early stage.

Most nuclear changes are accompanied by some form of chemical change. An isotope of another element may be produced, an atom may recoil into the surrounding medium where it may react so as to appear in a new chemical form, or some chemical change may be brought about in the surrounding medium by means of the radiation emitted. In the late 1930s, a great many radioactive isotopes had been produced by exposure of the particular element to slow neutrons. When, however, the heaviest element, uranium, was bombarded by slow neutrons there appeared to be a number of products, none of which could be clearly identified. It was thought that the primary process would be the absorption of a neutron to give another isotope of uranium which would decay by β particle emission to give isotopes of one or more new elements beyond uranium. It was only when Hahn and Strassman, in 1939, identified one of the major products as an isotope of the element barium that it was realized that there had actually been a fission of the uranium nucleus.

It was from this point, of course, that atomic energy developed. All the fission products were chemically separated and their yield and radiation characteristics determined, then after it had been shown that neutron capture with subsequent β-ray decay did, after all, occur with uranium, a whole new series of elements beyond uranium was produced and studied by Seaborg and his school at the University of California. This actinide series extending from thorium, protactinium and uranium through neptunium, plutonium, americium, curium, berkelium, californium, einsteinium and fermium to mendelevium is of prime importance to atomic energy since thorium and uranium are the only naturally occurring sources of fissile isotopes of uranium and since protactinium, neptunium, plutonium, americium and curium are produced in irradiated fissile material in technically significant quantities.

Besides the actinide elements, the modern radiochemist can study the new elements technetium ($Z = 43$), promethium ($Z = 61$) and astatine ($Z = 85$) as well as polonium and actinium which were previously only obtainable in small amounts from uranium ores. Thus about fourteen new or virtually new elements have been made available for chemical investigation since 1939, making altogether perhaps the greatest accession to inorganic chemistry for over fifty years. Atomic energy, in its technical ramifications, has involved in some special way, about twenty other elements including the rare earths as a single group, but not including the elements used in ordinary materials of construction or in ordinary chemical reagents. Inorganic chemistry has, therefore, assumed a special importance in the development of atomic energy.

Whilst research in pure chemistry is essential to an industry based on the use of new or little known elements, developments in applied chemistry are required perhaps even more urgently than in orthodox industry, to bring ideas and processes to practical fruition. Schemes which may be quite simple as initially conceived cannot usually be made to work in a chemical plant without a great deal of detailed laboratory work to meet the exacting demands of the chemical engineer. Analytical problems particularly have always been especially severe in atomic energy where so many dangerous or expensive materials have to be accounted for.

From the point of view of applied chemistry, it is convenient to divide the field of atomic energy into the following branches: raw materials, production of fuel elements and

other special reactor materials, chemistry of nuclear reactors, chemical processing of irradiated materials and disposal and utilization of radioactive wastes.

RAW MATERIALS

During the war period the basic problem on the raw materials side was to produce uranium metal of an exceptionally high degree of purity in as high yield as possible from rich ores from the Belgian Congo. This was first achieved on the pilot plant scale in this country at the Widnes Laboratory of the General Chemicals Division of I.C.I. Ltd., and the present Springfields plant of the U.K. Atomic Energy Authority, was designed on the basis of this work. More recently, the exploitation of the much more extensive low grade ore deposits has been one of the major achievements in the atomic energy field. In general, quite simple leaching processes have been employed in the locality of the mine, using such common reagents as dilute sulphuric acid or sodium carbonate. The very dilute solution of uranium so obtained is treated chemically to recover the uranium. In the case of the sulphate leach, the liquor is passed through a bed of an anion exchange resin which holds the anionic sulphate complex of uranium and this may be afterwards eluted in a relatively pure, concentrated, form. This system, simple and elegant in conception, has required a great deal of work to bring it into efficient operation on a large scale. Its most spectacular application has been to the residues from the South African gold mining industry where uranium production now vies with gold as a major factor in the national economy. In Britain the work on chemical treatment of low grade ores has been centred at the Chemical Research Laboratory, Teddington. The group at this laboratory, which suffered a great loss recently with the death of its leader, Dr. F. H. Burstall, has been in existence since the war period when all atomic energy activities came under the Department of Scientific and Industrial Research.

FUEL ELEMENTS AND SPECIAL REACTOR MATERIALS

Turning now to the production of fuel elements, mention has already been made of the war time and immediate post-war work which led to the building of the Springfields uranium plant. Since then chemists and chemical engineers at the Springfields laboratories of the Research and Development Branch, with some support from Harwell, have developed new purification and reduction processes which, although similar in basic principle to the early processes, really represent radical changes in application. The new processes are very substantially cheaper than the earlier methods and recovery of uranium passing through the plant is practically quantitative. Production of the uranium metal billet and its fabrication into the finished, canned fuel element are essentially metallurgical and engineering operations. There are several special chemical operations associated with the production of fuel elements enriched in ^{235}U which have required a good deal of effort. For example, pure uranyl nitrate must be converted into uranium hexafluoride, the feed material for a diffusion plant and this introduces a whole new province of fluorine chemistry. To appreciate the progress which has been made it should be remembered that before the war, fluorine was regarded as an industrially rare and highly dangerous chemical. Uranium hexafluoride and the halogen fluorides were known only as highly corrosive and dangerous laboratory preparations. As a result of the war time atomic energy programme these materials are now used on a very large scale in routine industrial processes. The early development work in Britain was carried out in the Universities of Birmingham and Oxford, and by I.C.I. Ltd. Now the Research and Development Branch of the U.K. Atomic Energy Authority has its own laboratory associated with the diffusion plant at Capenhurst. A useful by-product of the early work was a series of new fluorocarbons, some of which, such as polytetrafluoroethylene (Teflon) and polychlorofluoroethylene (Kelef), have remarkable properties, and were made commercially available by the United States after the war. The diffusion plant operates smoothly and efficiently despite the unpromising materials which are used; the enriched UF_6 which is produced is converted either to a uranium salt or to uranium metal in a small chemical plant for subsequent blending or for direct use in reactor fuel elements.

A very large effort has been devoted to the production of special reactor materials such as heavy water, graphite, hafnium free zirconium, and so on. Although interesting chemical problems are involved, the main requirement here has been for chemical engineering and engineering development since there is already a good deal of basic physico-chemical information available in the open literature. The separation of hafnium from zirconium, for example, which is necessary on account of the high neutron absorption cross-section of hafnium, appeared at one time to be a formidable problem. Several quite simple methods have since been discovered however, both in the United States and in Britain, based on the use of solvent extraction, ion exchange, or chromotography so that from a chemical point of view this can no longer be regarded as a serious problem.

CHEMISTRY OF NUCLEAR REACTORS

Although the supply of materials has always been an important question in the atomic energy project, the reactor is naturally the centre of interest. The early production reactors presented few serious chemical problems. They operated at relatively low temperatures and the radiation conditions were such that few embarrassing chemical effects arose. The situation is quite different in the case of power reactors which are designed to operate at higher temperatures and usually at higher fluxes. Since the rate of chemical reaction is, on the average, about doubled for an increase in temperature of 10° C it is evident that the possibilities of reaction at 300° C are very different from those at 100° C. The reaction between the carbon dioxide coolant and the graphite moderator of the Calder Hall reactor is an example. At low temperatures, the thermal reaction is negligibly slow but it becomes sufficiently rapid above 400° C to give rise to the possibility of appreciable mass transfer of carbon in the reactor from hot areas to colder areas and it is probable that this factor alone will place a limit on the operating temperature of carbon dioxide cooled reactors. In addition to the thermal reaction, there is also a reaction induced by radiation. The rate of this reaction is proportional to the radiation flux but is practically independent of temperature, so that in addition to the limitation on operating temperature on account of the thermal reaction, there is also a flux limitation on account of the radiation reaction.

Power reactors with water moderators are usually designed to operate around 300° C at pressures between 1000 and 2000 lb/in^2 and corrosion under these conditions is much more serious than at low temperatures and pressures. It is therefore necessary to find the operating conditions under

which corrosion is a minimum and to develop a continuous purification and conditioning plant for the moderator. The aqueous homogeneous reactor in which the fissile material is dissolved in the moderator is to all intents and purposes an unusual form of chemical plant. Here a dilute solution of uranyl (^{233}U or ^{235}U) sulphate in heavy water is circulated through the core and the heat removed in an external heat exchanger. It has been found, at Oak Ridge, that the rare earth sulphates have a decreasing solubility with increase of temperature and at 300° C the solubility is so low that the fission products can be expected to precipitate on the finely divided insoluble corrosion products which will be present in the core fluid. This insoluble material can be removed from the system by means of a simple hydraulic cyclone or hydroclone. There are a number of difficulties in this system, of which the radiation corrosion of the core vessel is the most serious, but it possesses many unique advantages such as a good breeding gain for ^{233}U if a thorium oxide slurry is used in the blanket and the possibility of very simple continuous processing of the core solution.

As the operating temperature is pushed upwards, the number of chemical systems available to the reactor designer diminishes; liquid metals, fused salts or gas cooled refractories each offer a few possibilities, but all suffer from the disadvantage of a dearth of information concerning chemical behaviour even in the absence of radiation. No doubt some empirical solutions will be found to the problems which will be encountered but if progress is to be soundly based, a great deal of fundamental chemical work will have to be done in addition to a large amount of work in applied chemistry, metallurgy and ceramics.

CHEMICAL PROCESSING OF IRRADIATED MATERIAL

During the early years of the Atomic Energy Project, by far the biggest chemical problem was the development of a process for the extraction of plutonium from irradiated uranium.

The outline of the process now used in the chemical plant at Windscale was developed by a small team of chemists and chemical engineers working in the Chalk River Laboratories in Canada where some irradiated uranium and suitable facilities for radio active work were available. The laboratory work was done on a semi-micro-scale with a few milligrams of plutonium using the actual plutonium concentrations to be expected in the plant. The process was based on the principle of counter current solvent extraction and the solvent selected was dibutoxy diethylene oxide. In addition to demonstrating the suitability of the process for the extraction of uranium and plutonium from fission products it was also necessary to establish that the solvent was sufficiently stable to radiation of an intensity likely to be encountered in the plant. This was done by exposure to fission product solutions of various concentrations and finally checked by preparing a solution with activity of the same order as that to be expected in the plant. When the chemical ideas were sufficiently far advanced, pilot plants were constructed so as to permit the chemical engineers to study problems of plant design, whilst in the meantime, the main chemical effort was transferred to the United Kingdom. At this stage, the Industrial Group under Sir Christopher Hinton was obliged to take the bold step of beginning full scale plant design on the basis of these semi-micro-experiments at Chalk River. Responsibility for detailed development of the plant flowsheet was given to the Widnes Laboratories of General Chemicals Division of I.C.I. Ltd. The Widnes group concentrated on the behaviour of uranium which constituted the bulk of the material passing through the plant whilst the chemical engineering group at Chalk River provided information on the more active components, plutonium and fission products. Although much additional chemical work was done at Harwell on this primary stage of the process in which the uranium, plutonium and fission products were separated into three streams, a great deal more remained to be done on the final purification and reduction to metal. This work went ahead at great speed after the new radiochemical laboratories were opened at Harwell. Even so, research, development and plant design fitted into the prescribed timetable only by the narrowest of margins and all concerned experienced considerable relief when this novel plant worked smoothly and efficiently.

Chemical processing has often been regarded as an expensive bug-bear of atomic energy and reactor designers have frequently spoken of the need for some simple process which would obviate the need for costly and elaborate chemical plant. This argument appears to be all the more compelling when it is remembered that the processing requirements of a nuclear power programme are much less exacting than those of a military production programme. Decontamination factors could be reduced from 10^8 to 10^2 and separation of plutonium from uranium need not be quantitative. The difficulty about this is that low decontamination factors require the provision of shielding against radiation and protection against plutonium for all the subsequent operations of fuel refabrication and reloading into the reactor whilst the extra cost of full decontamination in a conventional plant is not large. Thus, in order to displace the conventional aqueous process, which is now fairly cheap, it is necessary to develop a different primary process which is very considerably simpler and cheaper. An attempt is being made to develop such a process at Harwell, in parallel with similar work in the United States and Canada. It has been found, for example, that when an irradiated uranium fuel rod is melted in a crucible, about 80% of the fission products concentrate in a thin skin of slag and it is possible that this very simple process might be utilized in special cases such as in connection with a fast reactor. Many other processes have been examined but it is significant that new chemical plants which are being designed by the Industrial Group for processing fuel elements from experimental reactors will be of the solvent extraction type.

DISPOSAL AND UTILIZATION OF RADIOACTIVE WASTES

We now come to an item which always appears last in an atomic energy list but which is by no means among the least, namely the disposal and utilization of radioactive wastes. I have described how the main chemical effort in the early years at Harwell was devoted to the chemical separation plant. The first thing that had to be done, however, before any radioactive work could begin, was the development of a radioactive waste system, and especially its analytical control, which was acceptable to the public bodies concerned. Similarly, one of the first things to be done at Windscale was to investigate the rate of dilution of an effluent solution in sea water, the movement of the sea water in bulk and the deposition of activity on the sea bottom, on the shore and in the local forms of marine life. Fortunately the chemical separation process adopted at Windscale permitted the main fission product waste solution to be concentrated by evaporation to small bulk and the product from many years of plant operation can be stored in tanks of moderate capacity. Thus there is no immediate problem although the permanent

storage of millions of curies of fission products in solution is not very desirable. The two most dangerous components of these old fission product solutions are ^{137}Cs, a γ-ray-emitter of half-life of 30 years and ^{90}Sr, a β-ray-emitter of half-life 28 years. If these two isotopes are removed, the remainder of the fission products decay sufficiently in 15–20 years to permit disposal into the sea or perhaps into the ground. A suitable process has been devised and production of ^{137}Cs has begun on a pilot plant basis at Windscale. It appears that all the caesium which is likely to become available during the next few years can easily be taken up either by hospitals or by industry as a source of γ-radiation. The market for the ^{90}Sr β-ray-emitter has so far only been developed on a small scale but there is no reason why this also should not ultimately be sold to industry in megacurie quantities. Even if the caesium and strontium cannot all be sold, it has been shown that they can be absorbed on special types of clays which can then be baked so as to render the activity practically completely insoluble. The baked radioactive clay could then be safely buried in the ground.

The availability of very large sources of radiation at a low price is of interest to the chemical and allied industries. Considerable work is going on in this field already and it is highly probable that radiation will become an important factor in the chemical industry.

The amount of radioactivity which can be released into rivers and lakes is naturally extremely small. On the other hand, the increasing occurrence of radioactivity in nuclear reactors, nuclear chemical plants or as radioisotopes, quite apart from nuclear explosions, undoubtedly constitutes a problem of increasing magnitude. There are two aspects of this problem. One concerns the disposal of large volumes of weakly radioactive effluents which are still far above the permissible discharge level to rivers and the other concerns the disposal of contaminated solids. A great deal of chemical and chemical engineering work has already been devoted to the development of distillation techniques, sludge precipitation methods and combustion plants for combustible solids. Recent work at Harwell has shown that a self-contained system based on the circulation of de-ionized water, which permits the efficient removal of radioactive contamination by a combination of sludge precipitation and ion exchange, might be a satisfactory solution.

I have described in this lecture, some of the chemical aspects of an atomic energy project, from the treatment of the ore to the final disposal of the wastes. The object has been to give an overall picture rather than to go into detail but I hope the picture has been sufficiently complete to give a satisfactory indication of the role of the chemist and his colleague the chemical engineer.

The role of metallurgy in a nuclear energy project

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The development of nuclear energy both for weapons and more particularly for peaceful applications is essentially a new branch of applied science. Like many new engineering projects some of the limitations to future progress depend on metallurgical difficulties. Problems arising in the construction of the large chemical plants for any nuclear power project are essentially the same in kind, although perhaps rather more acute in character, as those in normal chemical industry where dangerous and toxic chemicals are involved. On the other hand, the manufacture of fuels for nuclear reactors has required a new industry to be set up for the extraction of uranium, and other new metals are becoming increasingly important if progress is not to be retarded. Apart from new metals, new alloys and higher quality material in conventional metals have been required for the protection of uranium from attack by the coolants used. From the earliest days this has been one of the acute problems of nuclear energy. Finally the metallurgical problem peculiar to atomic energy is the development of materials which will withstand the effects of neutron irradiation. The effects on non-fissile materials are usually minor changes in physical properties, but on fissile material the effects are much more severe. At lower temperatures the anisotropic nature of uranium results in large dimensional changes which are being overcome by grain refinement techniques involving alloying and heat treatment. At high temperatures the gaseous fission products tend to expand and disrupt the material by mechanisms which are not yet fully understood, and for which cures are still being developed.

The position of the metallurgist in a nuclear energy project can be regarded in some respects as intermediate between the position of the engineer and that of the chemist. Where the chemist processes raw materials through to the point where a pure compound is produced the metallurgist takes over. He carries out the reduction process by one means or another to prepare metal in a suitable form. He manipulates the metal by a variety of processes and finally delivers to the engineer a finished product suitable for incorporation in his engineering device, which in nuclear energy is the nuclear pile. The problems of extraction and fabrication are, however, by no means all the metallurgist's problems. In any large engineering organization, such as the Industrial Group of the U.K. Atomic Energy Authority, quite conventional problems arise where the service of the metallurgist can be utilized. For example, work on corrosion or on the fabrication of chemical plants is just as essential in a nuclear energy project as in any other engineering undertaking. The only difference is that the chemical plants will operate under conditions where radiation (notably gamma radiation) will make inspection difficult, but in other respects the plants can be regarded as conventional. For the materials of the fuel containers for nuclear reactors, however, quite often we find that existing materials are inadequate for the duty they would be required to carry out, and even where conventional materials such as aluminium or magnesium may be suitable for service in the pile, it may very well be the case that metallurgical development is necessary to achieve the requisite quality for nuclear engineering purposes.

It is, of course, the fuel itself which provides the unique

metallurgical studies. The primary fuel materials are thorium, uranium and plutonium which are used either singly or in combination for the fuels of the Nuclear Power Programme. Each of these metals has its own distinctive characteristics, and for their most effective utilization, extensive studies need to be made both of their properties and alloying behaviour.

Finally, there is a new class of phenomena not encountered in any other branch of engineering and which is peculiar to atomic energy. This is the damage to materials caused by irradiation, which gives rise to a need for materials less susceptible to such damage than those available to us at the moment. We can, therefore, say that there are three types of problems which interest the metallurgist entering the nuclear energy field. Those of conventional engineering, those of the extraction of the fuel and of new metals for containing the fuel, and those of the development of materials suitable to withstand irradiation. Before considering these problems let us first look at the state of metallurgical knowledge and experience in our Atomic Energy Authority to-day.

EXISTING WORKS

We have built a number of major establishments—there is the Atomic Weapons Research Establishment at Aldermaston in Berkshire; the Atomic Energy Research Establishment at Harwell and the various works and laboratories of the Industrial Group. The Atomic Weapons Research Establishment, although its function is the manufacture and development of weapons, nevertheless handles exactly the same materials as are necessary in a Nuclear Power Programme, so that many of the investigations carried out in the Metallurgy Divisions at Aldermaston are similar to those in the Atomic Energy Research Establishment and in the laboratories of the Industrial Group.

The works which exist already in the Industrial Group consist of a uranium manufacturing plant at Springfields, a plutonium manufacturing plant at Windscale where a portion of the natural uranium is converted into plutonium by irradiation in the piles at Windscale, and a plant at Capenhurst where the two isotopes of uranium are separated. At Springfields where we are dealing with a more or less inactive material, natural uranium, the plant is relatively conventional. Precautions need to be taken, of course, in the chemical plants to avoid ingestion of uranium or its products such as radon, but the plant is accessible for maintenance and although great care has gone into its construction, it has generally been possible to use conventional materials. The chemical side of this plant has been dealt with by Dr. Spence in his lecture,* which also indicated similar requirements for the chemical plants at Windscale. Perhaps it is worth emphasizing again that since much of the Windscale plant is not readily accessible to inspection and maintenance because of the high irradiation levels, special care in the manufacture of the components and in their assembly has to be taken. At Capenhurst, where the gaseous diffusion process is employed, the main metallurgical problem which arises is that of corrosion of materials of construction by the process gas uranium hexafluoride. This was adequately dealt with in the early days of the project, largely in the laboratories of Imperial Chemical Industries Ltd. In the immediate future the Calder Hall plant will be operating. It will soon be followed by the Calder Hall B Power Station and soon after by the Chapelcross Station which again will reproduce the main features of the Calder Hall A Station. Many metallurgical problems have been encountered in the construction of these stations and in passing it might be worth mentioning that these have been essentially concerned with the construction of the pressure vessels, the production of the uranium bars which constitute the fuel in the reactors, and perhaps most of all with the magnesium alloys which provide the canning for the uranium bars. We shall soon have in operation our fast reactor experiment at Dounreay, in Scotland. In this case we are entering into a relatively new field of activity where very high heat production in a very small volume leads to acute metallurgical difficulties. The problem can only be met by the production of new materials and by the special treatment of our uranium fuels. Much effort has gone into these studies in recent years.

* See page S 88 of this Supplement.

FUTURE PROSPECTS

Turning to the future; in the longer term, we can see the prospect of new types of reactors coming along. Whether a water-cooled reactor or a sodium-cooled graphite-moderated reactor is a satisfactory device to succeed our gas-cooled reactor is not yet decided, but obviously one of these types or some alternative must soon be included in our future programme. Though we do not know what type will be selected we are quite sure of what the trend will be. The Calder Hall reactors are now subject to development by four teams of industrial designers which in each instance incorporate a heavy electrical engineering firm, a boiler-maker and other engineering skills. This arrangement greatly augments the engineering effort available for nuclear reactor development and although it is primarily set out for the development of gas-cooled reactors, no doubt other types of reactor will gradually be incorporated into the same system. In the case of the gas-cooled reactors which are being designed by these Groups at present, it is clear that not for long will they be willing to accept the limitation on thermal efficiency which arises from the relatively low temperature of operation of the Calder Hall reactors. This in its turn will lead either to engineering solutions of the problems involved, or to metallurgical solutions, or both. For instance, it is inconceivable that a very large increase in temperature of the fuel will be possible using aluminium or magnesium as a canning material. New materials must be developed and these could either be beryllium or zirconium, or means might be found for using steels or similar materials. There is no doubt, however, that the future will show a change from present practice.

CONVENTIONAL METALLURGY

Let us first of all consider the problems which arise in conventional engineering in a nuclear energy project. There is no need to spend a long time in dealing with this, but by way of illustration it may be worth while to refer to the work which has been done in connection with the chemical processing plants at Windscale. For normal plant handling corrosive materials such as those involved at Windscale, ordinary stainless steel would be used, since in essence the irradiated uranium rods are dissolved in nitric acid and are purified by a solvent extraction process. Ordinary stainless steel would have been expected to have a reasonable life in the circumstances envisaged, but corrosion studies indicate that the effects of process liquors containing materials with variable valency were not always those which were expected from laboratory studies, and detailed work indicated that stainless steel containing 18% chromium, 13% nickel and 1% of niobium was preferable to the more conventional 18/8 types.

Niobium was added as a stabilizing element for welding purposes in preference to titanium since in plant of this kind the amount of welding to be done was considerable and niobium was known to have advantages when a good deal of cross-welding was involved.

The plant as a whole needed to be constructed by welding, and demountable joints were not included since with a plant containing materials with a high γ-radiation level, it was never expected that the joints would be taken apart. It was felt to be impossible to maintain joints, and special techniques were necessary to ensure that they were of exceptional reliability. This may be regarded as a typical conventional metallurgical problem for the metallurgist in a nuclear energy project. It simply involves the selection of the most suitable materials of construction, the development of more precise welding techniques, and the provision of an adequate inspection standard to ensure that the quality of workmanship and finish throughout the plant was of the highest standard. Thousands of welds were made in the construction of the Windscale chemical plants and every weld was subject to its own individual inspection technique which depended on the service to be expected. Much of the work we have done has already been described in the literature and it has probably been of some value to workers in allied fields to our own.

Many other conventional metallurgical problems have been encountered as would be expected in large-scale engineering work. Each works introduces its own type of problem with the metallurgist acting as the assistant and consultant to the design engineer. This branch of metallurgical work is of great importance economically but on the whole, it is less interesting than the study of new metals which we encounter when we turn to the production of fuels.

FUEL DEVELOPMENT

So far the reactors which have been built have been heterogeneous in that the moderator and the fuel are separated from one another in the reactor. In this country the fuel has been in the form of metal rods; so far we have only used uranium as a fuel. Uranium is indeed the only available natural material for use in a nuclear reactor since it consists of the fertile material ^{238}U with the necessary admixture of ^{235}U, the fissile isotope, which is necessary for the chain reaction to proceed. It would be possible to use thorium to replace the ^{238}U and plutonium to replace the ^{235}U since the effect of neutron irradiation of thorium would be to produce ^{233}U and plutonium is itself a fissile element like ^{235}U. Of the possible fuel materials uranium is perhaps the most interesting. It is a metal of high density but of low crystal symmetry. The density is useful but the lack of symmetry in the crystal adds to the difficulty of using it. This will be discussed later when we consider the behaviour of the fuels under irradiation. Plutonium itself is considerably less convenient to use than is uranium. Whereas uranium has three allotropic forms, plutonium has six and melts at a low temperature compared with the melting-point of uranium. It is perhaps rather doubtful whether plutonium could be used in metallic form in a reactor except when alloyed with other elements whereas uranium has already been used in un-alloyed form with great success. Thorium, on the other hand, unlike uranium, is free from allotropic changes and has a cubic symmetry, thereby being a relatively isotropic material. It might be expected on this account to be less susceptible to irradiation damage than uranium.

Let us consider the effect first of the variability of properties in uranium according to the crystallographic direction. In uranium the crystal lattice is orthorhombic; the coefficient of expansion in one direction in the crystal is high, in another direction it is low, and in the third direction it is negative. Consequently on heating and cooling, quite a considerable thermal stress is set up which may lead to distortion of the bar of metal used for the fuel element. The effect of irradiation is to some extent similar. At 200° C a single crystal of uranium would about double its length with a corresponding reduction in its diameter for a burnup of one in a thousand atoms. The effect of this on cast material of large grain size is that it exhibits severe roughening or wrinkling of the surface, while rolled metal increases in length and decreases in diameter or vice versa according to the rolling temperature. Powder compacted metal has been known to do both and this distortion may lead to stretching of the can in which the uranium is enclosed and in some cases to actual rupture of the can. A further complication arises if one should allow the uranium to pass through the alpha-beta transformation which occurs at about 650° C. Repeated heating and cooling of uranium through this temperature in the absence of irradiation will lead to bars which distort in length and in diameter, or in which very badly roughened surfaces may be produced. The metallurgist's problem is to reduce these effects to a minimum or to eliminate them. Where the effect is the property of a single crystal, the distortion is greater for large grain sized material than for small, and a palliative is found by heat treating the uranium to give a smaller grain size, but if the effect is not associated with the lack of symmetry in the crystal some other solution must be found.

What are the effects which may be encountered which are not associated with this lack of crystal symmetry? Obviously during fission the fission products formed may or may not be soluble in the uranium; clearly the alloying properties of uranium are of interest here. The fission products may be gaseous, and if these can diffuse through the uranium and accumulate at suitable nuclei to form gas pockets, then bubbles of gas will form within the solid metal. The effect of this will be to cause an overall volume expansion in the uranium. The extent to which the gas bubbles will grow will depend on the restraint imposed either by the uranium itself or by the container in which it is held. If we attempt to make the swelling as small as possible by increasing the strength of the uranium we are obviously concerned at once with research into the effect of alloying on the high-temperature strength of uranium, but we have a limitation which is not normally imposed on the metallurgist responsible for developing high-temperature alloys in conventional metals, in that our alloying elements must be chosen from those with very low neutron absorption cross-sections, otherwise only very dilute alloys can be used.

We can summarize the work which has been done on the metallurgy of uranium, therefore, into a number of categories. Once the pure metal has been manufactured, new fabrication techniques have had to be devised, in part because of the high reactivity of the metal. An example would be the use of vacuum casting techniques to supersede the earlier air melting and casting techniques which were used. Following on the fabrication we are interested in the study of the heat treatment of uranium both within any single phase region or on passing through the phase transformations, and finally we are concerned with the development of new alloys with improved properties.

CANNING

An alternative means of preventing distortion in the uranium fuels is to enclose the uranium in a strong container

or can, but this container has an additional function in protecting the highly reactive uranium, or thorium if thorium be the fuel, from its environment. Generally speaking, this means protecting the fuel from attack by the heat transfer fluid whether it be gas or liquid. Air is used as a coolant for the reactors at Windscale, while in the Calder Hall reactors carbon dioxide has been used. With the gaseous heat transfer fluids it is necessary to provide an extended surface on the can so that an adequate amount of heat can be taken away by the fluid. This would not be necessary if water or liquid metal were used as a coolant. At Dounreay we are using liquid sodium as coolant for the fast reactor, but even in this case it is necessary to provide a suitable container to protect the uranium and to prevent it deforming excessively. As far as possible, of course, one would wish to use metals which are readily available and one assumes well understood. For this reason in our gas cooled reactors so far aluminium and magnesium have been used.

At Windscale, where air is used as a coolant, aluminium has been selected as a suitable canning material. Since the piles are primarily plutonium producers the time spent in the pile by the uranium is more restricted than would be the case if power were the objective. Consequently the distortion likely to be encountered is a good deal less and the strength of the container is of less importance. At surface temperatures which are up to about 400° C aluminium is not, of course, strong and the primary purpose of the can in this case is to protect the uranium from the very rapid attack which would be encountered if it were exposed to the cooling air. It might be thought that to use a well-known material such as aluminium required very little development effort, but in fact we have had to work on many aspects of aluminium technology, including the melting of the original material. In this we have had excellent collaboration from some branches of the aluminium industry. The manufacturers have by careful selection of raw materials and careful control of the melting procedures, produced an aluminium of commercial purity which is undoubtedly cleaner than any ever produced for other purposes, and this has proved a very satisfactory material for us. Again, great care has had to be taken in the sealing of the cans using argon-arc welding. We have ourselves worked on this technique and on suitable inspection methods to ensure that good results are obtained. Finally, since a finned can is necessary for adequate heat transfer, we have, again with the collaboration of branches of the industry, carried on over the years much development work on the extrusion of aluminium, and to-day the extruded cans which are produced for our purposes might very well be regarded as model extrusions; they are of the very highest quality and probably excel anything made for other purposes.

The Calder Hall reactors are cooled by carbon dioxide and it was felt worth while in this case to examine whether it would be possible to use magnesium as an alternative to aluminium as the canning metal. The first doubt which had to be dealt with was on the score of inflammability. An investigation carried out at Harwell in collaboration with the magnesium manufacturers indicated that it would be possible to produce suitable alloys of magnesium which would be free from any serious risk of burning in the pile. Subsequent work has shown that for the operating conditions in the pile no great hazard would be encountered if commercially pure magnesium were used as an alternative. At the same time investigations were started at Harwell on the production techniques which would be necessary to use magnesium in this way. It was established that there was a good chance of using magnesium, and it was therefore selected as the canning material for the Calder Hall reactors and is likely to be used in the first industrial power reactors which will be built in the next few years. During the design and construction stage of the Calder Hall reactors, a great deal of development work has had to go on into the manufacture of high-quality magnesium, and into modifying the alloy composition to ensure good machinability and reliable welding quality, until at the present time very satisfactory materials are available for this highly important duty. Magnesium has the advantage over aluminium that with a lower neutron absorption cross-section, rather thicker sections can be used, and with a slightly higher creep strength of the metal, some restraint of the uranium is possible. This will probably be of benefit in restraining the distortion and growth of uranium which has been referred to earlier. On the other hand, magnesium is less ductile than aluminium and some doubts have been expressed as to whether it possesses adequate ductility to allow for some distortion in the can before rupture begins. We are confident, however, that in the Calder Hall reactors no difficulty will arise on this account.

It is when we turn to the future that this canning problem, which in many respects is the central problem in the metallurgy of reactors, becomes most interesting. In the case of the Dounreay fast neutron reactor, temperatures of the uranium are higher, the consequent distortion of the uranium due to gas pockets forming will be greater, and it will be almost impossible to ensure that the uranium does not pass through the alpha-beta or even the beta-gamma phase transformation. The probability is, therefore, that distortion will be greater and the need for a strong canning material becomes progressively more important, not only in this case but in the case of any of the more highly rated reactors which the future will bring forward. We have, therefore, to consider the development of new metals with higher strength than our characteristic canning metals—aluminium and magnesium—for future service. Obviously the first material which one thinks of is steel, and steel would have some greater use in reactors if it had a lower neutron absorption cross-section and if iron and uranium did not form a low melting-point eutectic. This low melting-point eutectic means that the compatibility of uranium with iron or nickel based materials is low. Consequently we look ultimately to the use of new metals such as beryllium, zirconium, niobium, vanadium, tungsten, molybdenum and so on.

Where neutron absorption is a vital factor as in the thermal neutron reactors, beryllium and zirconium have some real advantages. Beryllium has a lower absorption cross-section than magnesium and aluminium, and it has better high temperature properties. On the other hand, it has only been made in small quantities in the past and it is known to have very low transverse ductility, and indeed some doubts have been expressed on whether it will ever have good ductility. It is also difficult to work and difficult to weld. A great deal of work, however, has been done both at Harwell and at Aldermaston on the fabrication and testing of beryllium, and we feel that it has been established that the metal could be used effectively in thermal neutron reactors, but since the price of beryllium will remain high, the conditions under which it should be used in substitution for magnesium and aluminium need very careful thought.

The alternative to beryllium for high-temperature service is zirconium. Zirconium has been produced in greater quantities and the amount of development work which has been carried out on zirconium is greater than in the case of beryllium. There is little doubt that the high-temperature strength will be increased, but the resistance to attack by air

and by carbon dioxide leaves some doubts as to whether it can be used at very high temperatures. Zirconium has an additional possible use both in sodium-cooled reactors and in water-cooled reactors, and especially in the latter case it is probable that it would have advantages since it is likely to have a high resistance to attack by hot water.

If we consider materials for reactors cooled by liquid metals we find that beryllium unfortunately is less resistant to attack by liquid sodium than some other metals; aluminium is very rapidly attacked; magnesium is unsuitable and there are some doubts as to whether zirconium would have sufficient strength if the temperature is raised appreciably above present levels, so that for reactors operating at high temperatures with liquid metals as coolants we look to alternative refractory metals for possible canning materials. Two which have been developed extensively in this country are niobium and vanadium. Fortunately, both these metals were made in small quantities in this country and the development work on extraction and on fabrication was able to begin at once with metal which could be obtained fairly rapidly. The general quality of the material available was not as good as we would ordinarily require and in collaboration with the manufacturers of the metal we have ourselves carried on very extensive development programmes. Niobium in particular shows very great promise. It appears to be amongst the most resistant metals to attack by uranium and indeed it is possible to hold liquid uranium in niobium crucibles for quite appreciable times. It is also extremely resistant to attack by liquid sodium especially if the sodium can be kept free from contamination by oxygen or nitrogen. Niobium additionally in the pure form is an extremely ductile metal and can be readily fabricated into sheets and tubes. Its high temperature strength even so is good and it appears to be a metal capable of very ready development into alloys with very favourable high-temperature properties. This combination of advantages would suggest that in certain types of reactors niobium would be the preferred canning material. The neutron absorption cross-section unfortunately is fairly high which would limit the amount of niobium which could be used in a thermal neutron reactor. However, its extremely good ductility enables it to be drawn down to very thin tubes and sheets which allow some use of niobium even in thermal neutron reactors. The fast neutron absorption of niobium is less unfavourable and our own immediate use of the material is likely to be in a fast neutron type of reactor.

The position with vanadium is much the same as for niobium. Much development work has been needed to produce suitable materials for reactor use, but there is little doubt that it will be available when needed.

It will be seen that in connexion with these newer materials a whole range of problems lie before us. Up to the present we have been busy ensuring that suitable extraction and fabrication techniques are available to provide the pure metal in suitable shapes, but this work will need to be extended to give materials with a wider range of properties. In particular we shall require to develop alloys.

CONCLUSIONS

The part played by metallurgy in the U.K. Nuclear Energy Project has been varied. By far the most interesting work has been in the extraction and development of new materials whether they be fuel materials or new metals for canning. This work is by no means at an end and will undoubtedly be extended. We can anticipate that heat treatment and alloying of our newer materials will become increasingly necessary and that metallurgy will remain an important branch of nuclear power development for many years to come.

SESSION 8. DISCUSSION ON "RESEARCH REACTORS AND THEIR USES"

Chairman: T. E. ALLIBONE, D.Sc., M.I.E.E., F.Inst.P., F.R.S.

The Canadian research reactors and their uses

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The heart of the programme at Chalk River is the high-flux, heavy-water-moderated NRX reactor. The primary aim justifying this and the even more costly NRU reactor is to explore the possibilities of deriving low-cost electricity from nuclear energy. This aim is not, however, pursued narrowly and the reactors are used to foster basic research in nuclear physics, nuclear chemistry, radiation physics, chemistry and biology. Since separated fissile material has high intrinsic value in atomic energy research, emphasis has been and still is given to the production of plutonium and uranium-233 as well as to certain radioisotopes, tritium and cobalt among others.

Nuclear power will become important within Canada only if it is cheaper than power from the present-day alternatives. The more costly of these are coal in Ontario, coal and oil in the Maritime Provinces, and oil in remote industrial installations such as mines and wood-pulp mills. Coal in Ontario at $8/ton and contributing 3 mills/kWh to the cost of electricity, sets the major target. Results obtained in the NRX reactor indicate that once the large development costs have been worked off nuclear power costs will be definitely lower than this target.

Canada is relatively rich in uranium and must immediately consider the particular requirements of foreign as well as domestic consumers. Consequently the research remains broad, and very full programmes lie ahead, not only for the NRX reactor, but also for ZEEP which is mostly used for measurements on fuel arrangements, and a proposed PTR ("Swimming-Pool" Test Reactor) which will take over the function of ZEEP for measuring reactivity effects of materials, particularly after irradiation. The most powerful and versatile research reactor is, however, the NRU, a heavy-water-cooled and moderated, high-flux reactor, which should be in operation in 1956.

From the beginning, the heart of the Canadian Atomic Energy Project at Chalk River has been the high-flux, heavy-water-moderated NRX reactor (Fig. 1). Until 1952 this held pride of place among research reactors for the highest neutron flux, about 6×10^{13} n cm^{-2} s^{-1}. Then it was surpassed by the United States Materials Testing Reactor at Arco, Idaho, which made available for the irradiation of small specimens a flux as high as 4×10^{14} n cm^{-2} s^{-1}. At the beginning of 1951 we started to design and construct a new heavy water moderated reactor that should give about five times the flux of the NRX. This new reactor (Fig. 2), the NRU, is a very large project costing close to $50 million and we are hoping that it will come into operation before the end of this year.

Our primary interest has been to explore the possibilities of deriving low cost electric power from nuclear energy, but we do not pursue this aim too narrowly. We realize that our knowledge of nuclear phenomena is still rudimentary and that nuclear physics, nuclear chemistry, radiation physics, chemistry and biology all offer challenges for basic research. The basic research scientist at times protests that fundamental scientific research is such a proper way of life that it needs no excuse, but in practice work on the nuclear science frontier demands so much co-operation from industrial civilization that much time and careful thought has to be given to choosing a research that will reward not only the scientist but also the whole supporting organization, which for a government supported project includes every taxpayer. It is the lot of those responsible for such projects to spend much of their time correlating the ways and the means. Certainly in Canada, as in most countries, any expenditure such as the $50 million we are spending on the NRU reactor would not be in the realm of practical politics if it were merely a means of scientific research. Our NRU reactor therefore has to earn two-thirds of its cost and keep as a producer of plutonium and radioactive isotopes. Even the remaining one-third would not be considered justified by basic research, which from the point of view of direct financial return to the backer must be considered a risky venture, because essentially the outcome must be unexpected. In the narrow field of nuclear power development, however, we can see the justification.

In Ontario the large-scale reward should come soonest, but before the end of the century every province in Canada seems likely to benefit directly from nuclear power, and the indirect benefits will be shared much sooner. The direct return in Ontario arises before 1970. The Hydro Electric Power Commission of Ontario this year presented a brief to the Royal Commission on Canada's Economic Prospects. This forecast that during the 1960's most of Ontario's new electric power requirements would be met by constructing generating plants burning fuel. If this were all coal, then at $8/ton by 1970 the annual fuel bill would lie in the range $100 to $130 million. By the use of uranium instead, we at Chalk River see the possibility of saving two-thirds of such a fuel bill. This goal justifies the $18 million capital expenditure on the NRU reactor and a similar sum on the nuclear power demonstration reactor, NPD, that we are also designing.

This prospective fuel saving, although attractively possible, does demand that we should choose our research programme with care. It is my personal belief that the lines we are following are highly promising and can stand comparison with any reactor development programme in other countries. I still hope that others will also adopt a route closer to ours, for there is plenty of room. It is characterized by the very long irradiation of solid fuel having a substantially constant

power output. This is made possible by paying great attention to economy of neutrons and using heavy water as moderator with this object.

THE LONG IRRADIATION OF CONSTANT POWER FUEL

No direct experimental work on this has been reported other than carried out in the NRX reactor. Four papers presented at the Geneva Conference last year referred to this work. In particular, D. G. Hurst[1] discussed the way in which the NRX reactor was itself adapted to make highly sensitive measurements of the reactivity retained by the normal fuel rods after an irradiation yielding 3000 MWD/ton. The results indicated less reduction of reactivity than predicted. This is encouraging because it indicates that no

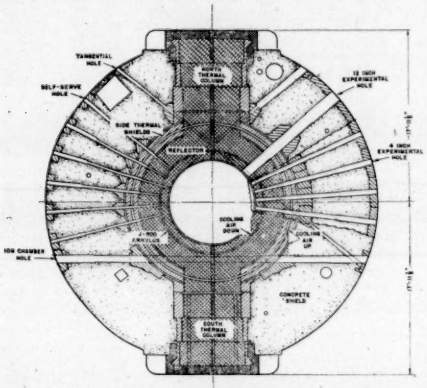

Fig. 1. The NRX reactor

serious poisoning arising from such an irradiation had been overlooked. The results were supported by a very detailed examination of a selected fuel rod after a long irradiation. Sections cut from this rod were examined for reactivity by the pile oscillator technique at the Argonne National Laboratory in the United States and the results were reported at Geneva by Spinrad, Carter and Eggler.[2] Other measurements of the distribution of plutonium isotopes and fission products in this rod will be published in due course.

By the irradiation of enriched fuel it is possible in a shorter time to attain energy yields of many thousands of megawatt days per ton. These irradiations are very useful for discovering the physical stability of fuel elements when the fission product concentration becomes high. Moreover, the irradiations can be carried out under conditions of temperature and thermal gradients appropriate to fuel elements in power reactors. Now that we have access to sufficient uranium enriched in ^{235}U, we expect to increase the number of such experimental irradiations in NRX.

Neither of the types of irradiation just mentioned provide the needed information on the reactivity changes of fuel in reactors in which the neutron temperature is high. Most unfortunately it is now established that the properties of all three of the plutonium isotopes ^{239}Pu, ^{240}Pu and ^{241}Pu are highly sensitive to neutron temperature and the neutron yield from ^{239}Pu decreases significantly at a high temperature. This has led us to emphasize power reactor designs in which the heavy water moderator is kept at a low temperature, and only the coolant takes the high temperature needed for efficient power generation. We hope to obtain the needed practical experimental information from our NPD reactor, in which the temperature of the moderator and coolant are independently controlled.

A number of papers presented at Geneva referred to the superior neutron economy promised by the ^{232}Th–^{233}U cycle. It appears, however, that this advantage is only to be expected in neutron fluxes below about 10^{14} n cm^{-2} s^{-1}, because at higher fluxes the intermediate product ^{233}Pa of 27-day radioactive half-life is significantly destroyed by neutron capture. The NRX reactor has a high enough flux to be well suited for studying this fuel. It appears possible to start with a mixture of ^{235}U and thorium that will pass progressively over to ^{233}U and thorium while maintaining a fairly constant power output in a constant neutron flux. We have an experimental programme for irradiating various mixtures of ^{235}U, ^{233}U and thorium to study the reactivity changes in collaboration with Staff at Harwell where pile oscillator measurements are made with the reactor GLEEP. In the near future we expect to resume pile oscillator experiments at Chalk River using a new swimming pool type of reactor, specially designed for such experiments with irradiated fuel elements.

SHEATHING AND STABILITY OF FUEL ELEMENTS

Next in importance to the long irradiation reactivity experiments, are those testing fuel under power reactor operating conditions for physical stability and survival following the development of defects. In this field we have had since 1950 a co-operative programme with the Westinghouse Atomic Power Division. It was through these experiments that we have been led to a preference for uranium oxide rather than uranium metal for the fuel. This preference is based on the effective immunity from chemical action with hot water, but it also appears capable of withstanding a longer irradiation than the pure metal without excessive deformation. Actual experience cannot be fully discussed, but, both in the metal and the oxide, there is need for scientific understanding of the behaviour of the fission products and we have a programme directed to this object.

One problem concerns any pressure exerted by the fission product gases inside the sheathing. It may be noted that about 30% of all fission products that are stable or have a long radioactive half-life are isotopes of the gases krypton and xenon. The most common textbook view is that these gases are trapped in intergranular defects and voids and there accumulate to build up a considerable pressure. At an irradiation of 4000 MWD/t, the gases would exert a pressure of 1 atm if they were free to occupy the whole volume of the oxide, and it might be supposed that if the void space is small and the temperature high, the pressure would be several hundred atmospheres. Direct evidence for this is lacking except when the fuel is near its melting point and the following indirect evidence suggests that this view may need considerable modification. Dr. A. H. Booth sealed up some zirconium metal powder in a capsule with argon. After irradiation in the NRX reactor a significant amount of argon was found to be firmly attached to the zirconium. Although most was released on heating to between 60 and 100° C, a small amount was retained up to several hundred degrees centigrade. There appears, there-

fore, to be at least some action in reverse to the supposed struggle of rare gases to escape. Many other observations might be quoted in support, but I am not aware of any work that affords a detailed scientific understanding of the phenomenon. In addition to a general exploration, we are planning from the escaping fission products, or for specific fission products. When the coolant is heavy water, the presence of photo-neutrons complicates the identification of delayed neutrons, but it still proves possible. When the coolant is pooled and recycled, there is a rapid spreading of radioactive

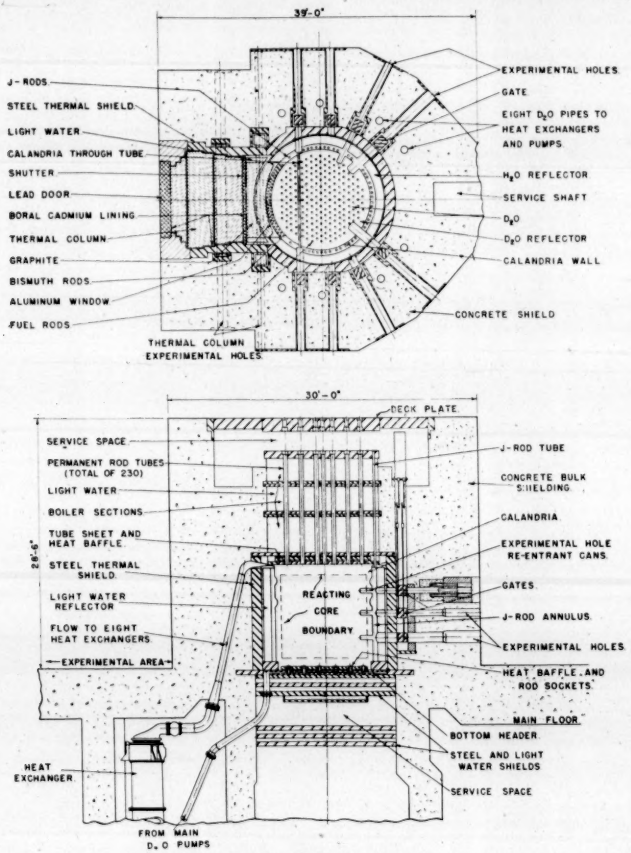

Fig. 2. The NRU reactor

also a direct experiment to measure the gas pressure inside a capsule almost filled with high density uranium oxide under irradiation in NRX.

Following any development of a defect in the sheathing, we wish to locate the fuel element affected, and therefore wish to have a means of detecting a small escape of fission products. The conditions are simplest in the NRX reactor where the coolant is ordinary water and is not recycled; monitoring can be for total γ-radiation, for delayed neutrons contamination from any one fuel channel to the others. It appears best to rely on changes in the operation of the reactor to provide a pulse of high activity from the defective channel in order to identify it. It would naturally be the objective in a power reactor to reduce the occurrence of any such defects to a minimum, but economic considerations may limit the allowance for fabricating and testing the fuel, so it seems necessary to continue to develop the means of monitoring for sheathing defects.

NEUTRON ECONOMY—LATTICE EXPERIMENTS

Except for a three-year interval from 1947–1950 when there was not enough heavy water, we have kept the ZEEP reactor, a zero energy heavy water critical assembly, continuously employed on measurements related to neutron economy in the design of reactors. When restarted in 1950 our main concern was with the design of fuel element and lattice for the NRU reactor. The basic design for NRU had been evolved in 1949. The objective was to build a research reactor using only natural uranium as fuel to achieve a much higher neutron flux than in NRX. Developing from NRX it would be necessary to increase the uranium surface and the amount of coolant. It appeared that using ordinary water as coolant the flux could not quite be doubled, while maintaining sufficient reactivity. Moreover, at the higher flux it would be desirable to retain a higher margin of reactivity to reduce the period of poisoning out by ^{135}Xe after shutdown. In NRX the period lasted from about 40 min after shutdown for about 16 h. With NRU it is expected to be 48 h. For these reasons it was soon decided to turn over to heavy water coolant.

We had found in NRX that the fuel rods remained very straight under irradiation, and this told us that we could hope similarly to constrain long thin flat rods of uranium, which would have the greater cooling surface required. This was soon confirmed by testing such rods in NRX. A fuel assembly consisting of five such rods in a surrounding tube was proposed. We could not, however, feel confident about calculations of the reactivity of lattices composed of such fuel assemblies. Lattices were therefore assembled in the ZEEP reactor as soon as enough fuel elements could be made. These experiments gave us the basic data for the NRU lattice. It was, however, also necessary to obtain a measure of the temperature coefficient of reactivity, and later to study the complex effect of a number of control rods in the lattice; for these the ZEEP reactor proved most valuable.

Turning attention to possible reactors for economic power, it became of interest to see whether distributions of fuel in other than uniform lattices would offer any economic advantage. Postulating a need for fuel processing set by physical deformation rather than from loss of reactivity, it was reasonable to examine whether a reactor could be made smaller in overall dimensions by enclosing the core within an absorbing blanket of depleted uranium or thorium. The properties of close-spaced lattices have therefore been studied, but it appears that the blanket cannot offer much reduction in diameter and requires many tons of uranium. To make closer estimates of smaller changes in lattice pitch, a double approach is being made. An analogue computer with a capacity of twenty lattice points is being used, checked and supported by a small number of measurements in ZEEP.

The swing of interest towards oxide fuel has set up another complete programme for measurements in ZEEP.

We have also made up a set of plutonium fuel elements for exploring in ZEEP not only the effects of plutonium growing in the fuel elements but also the application of recycled plutonium as fuel elements for power reactors.

REACTOR CONTROL

We propose in the NPD reactor to avoid the use of control rods altogether and to base all rapid control on the level of heavy water. This possibility and the appreciation of its merits arise from our experience with the NRX reactor. Originally the NRX was equipped with four control rods as well as with means for adjusting the heavy water level. For many years, however, we have used the heavy water level as the main control and only one control rod automatically operated for fine control. This has proved quite satisfactory for control except when using the reactor to make measurements that depend on the flux distribution. This is distorted inconveniently by the control rod. In a few months we shall be changing the method of control in NRX to a system in which ports are always open for the heavy water to run down to a prescribed low level, but a higher level is maintained by pumping heavy water in at a rate controlled by the neutron flux monitoring circuits. This will give experience that should be closely applicable to the system proposed for NPD.

GENERAL OPERATING EXPERIENCE

One of the main uses of our heavy water research reactors has been to provide experience. Some of the most valuable experience has been at the time the most painful. We have learnt where economy is unwise. In general it has proved important to exercise forethought for meeting all situations in which it is necessary to deal with high radiation fields and high levels of radioactive contamination. At the same time it has proved unwise to accept complicated mechanical designs. A mechanism that fails when handling highly radioactive material can prove the greatest embarrassment. We have discarded many devices for handling irradiated capsules but have still not achieved a perfect device.

Our major accident of December 1952 found us well prepared to save most of the heavy water, but quite unprepared to deal with a million gallons of highly radioactive water flooding the basement. There was no prepared place to take the water, and as it stayed in the basement it seeped deeply into the unprotected concrete. In the reconstruction we have provided surfaces that are relatively easy to decontaminate and this has subsequently proved most helpful.

The lessons learnt concerning reactor design, control and safety measures, have been described elsewhere and involve the rather subtle principle that shut-down circuits should always take effect. This means that as soon as possible after a shut-down, some shut-off rods are withdrawn so that they are available to take effect if unexpectedly needed.

OTHER USES

Within the compass of a single paper it is only possible to touch on a few highlights of the other work, in fact it will be necessary to pick only a few for brief mention.

A study on the processes of fission has been maintained, dealing with the spread of energy and nuclear mass of the fragments, the angular distribution of the neutrons, the light nuclear charged particles emitted in fission, and the differences between fission of the many fissile nuclei.

Beams of neutrons have been used for the many studies possible with neutron spectrometers. Neutrons have been used for determining by diffraction the crystallographic texture of thick samples of uranium metal, a study not possible by other means because of the strong absorption. Diffraction studies have also been made of liquid helium, liquid oxygen, argon and nitrogen, as well as of organic molecules. Changes of neutron energy on scattering reveal the motions of nuclei in crystals. The magnetic interactions of neutrons reveal the antiferromagnetic structure of some crystals. This general field of solid state studies by the diffraction of neutrons promises to be highly useful to industry.

Two pioneer studies of nuclear energy levels have been made by measuring the γ-rays resulting from neutron capture in most of the elements. One of the first of these disclosed the significant error in the supposed mass of the neutron.

The classic experiments of Robson on the radioactive decay of the free neutron should also be mentioned. Most recently he has studied the energy and angle correlation between the decay electron and the proton. This has confirmed the applicability of the theoretical interpretation and has pointed up as most important a more precise determination of the radioactive half-life.

The high flux of the NRX reactor has been applied not only to make radioactive isotopes such as ^{60}Co with high specific activity (20 c/g) and amounting to tens of thousands of curies, but also to make isotopes resulting from two, three and more successive captures of neutrons.

A high flux of fast neutrons is obtained inside a tube of uranium with a wall thickness of about a centimetre which we call a neutron velocity transformer. This has been used for a number of studies on energy storage in graphite, order-disorder transitions in alloys, and changes of other physical characteristics such as resistivity.

Neutron deficient isotopes such as ^{22}Na have been made by the action of tritons expelled in the disintegration of lithium following neutron capture.

Measurements of effective cross-sections in different types of neutron flux are made. A mainly thermal flux is available in the graphite thermal columns, also in a vacant lattice site. A higher proportion of fast neutrons is available adjacent to fuel elements, in a neutron velocity transformer or under a cadmium shield.

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The United Kingdom research reactors and their uses

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A brief description is given of the U.K. research reactors. Five are in operation at present and four more are in process of being constructed. They are:

(1) GLEEP, a low flux natural uranium graphite-moderated reactor;
(2) BEPO, a medium flux natural uranium graphite-moderated reactor;
(3) DIMPLE, a low flux heavy-water-moderated reactor, designed to take a variety of core arrangements.

The remaining two reactors already operating are fast reactors. These are:

(4) ZEPHYR, a plutonium fuelled reactor and
(5) ZEUS, a ^{235}U fuelled reactor.

The four reactors being built are all thermal reactors. Three use heavy-water moderation, and are designed to give a flux of 10^{14} n cm^{-2} s^{-1}. The fourth is natural-water moderated of the "swimming-pool" type, and should give a flux of 10^{12} n cm^{-2} s^{-1}. These four reactors will all be fuelled with uranium 235.

The uses of these reactors are listed and a description of some of the more important uses is presented. In particular the effect of radiation on chemical reactions is receiving considerable attention.

In the reactor technology field, work in high-flux reactors is essential and this is taking the form of so-called "loop tests." These are small sections of proposed reactor cores built into research reactors and cooled by a separate circuit.

A "family tree" can be drawn up of the nuclear reactors so far built or planned for building in the United Kingdom. In this tree, distinct branches can be traced, and though this paper will not pursue it, it would be fascinating to consider the potentiality of reactor systems resulting from inter-marriage between the branches.

The branches are

1. The gas-cooled, graphite-moderated natural uranium reactors represented by the sequence GLEEP, BEPO, Windscale and Calder Hall. This branch can trace its ancestry back to Oakridge, Tennessee.

2. The heavy-water moderated reactors (with earlier links in both the U.S. and Canada) comprising DIMPLE, DIDO and PLUTO.

3. The natural-water moderated reactor LIDO, with close family ties in U.S.A.

4. The fast reactors, represented by ZEPHYR, ZEUS and the Dounreay reactor, now being built. This branch of the family is completely British.

It is the purpose of this paper to describe briefly the research reactors among these mentioned and to indicate their uses. A more detailed description of the reactors is given in reference (1).

GLEEP

Reactor GLEEP is an air-cooled, graphite-moderated reactor using natural uranium fuel. It is essentially a 24 × 24 × 21 ft stack of graphite blocks with horizontal channels into which lengths of uranium are inserted. In the central region the uranium is in the form of metal bars, 0·97 in. in diameter in various lengths loaded to fill the central 17 ft of the channels; 12 t of metal is used. Outside this region, uranium oxide, pelleted to a density of 6·5 g/cm^3 and slipped into thin-walled aluminium tubes 1·6 in. in

diameter, is loaded to make the system slightly above critical size. The total weight of uranium oxide is 20 t and the loaded diameter of graphite is 19 ft. The graphite stack is surrounded by 5 ft of concrete on all sides to act as a biological shield. Air can be blown through one end of the shield, over the uranium to cool it and out through a hole in the roof. During the early life of GLEEP this cooling system was used to remove 100 kW of heat, but since this heat-output was exceeded by reactor BEPO, GLEEP has been restricted to a heat output of a few kilowatts only.

This heat output gives thermal neutron fluxes of the order of 10^9 n cm^{-2} s^{-1} at the centre of GLEEP, which is adequate for the main use to which it is put. This main use is in comparing the neutron absorption and production properties of materials. An absorber placed in the centre of a pile will cause its flux level to fall. If the absorber is withdrawn from the pile the flux level will rise again. If the absorber is oscillated in and out of the high flux regions of the pile in some regular fashion, the flux level will rise and fall with an amplitude of oscillation which depends on the neutron absorption in the sample. By suitable calibration, therefore, it is possible to derive absorption cross-sections or compare nuclear purity of samples of materials. Conversely, if the sample oscillated produces neutrons, e.g. a Ra–Be source or fissile material, then the amplitude is a measure of the net yield of neutrons from the sample.

The advantages of the method are first that with suitable equipment an extremely high sensitivity is possible. GLEEP can observe an absorption cross-section of 0·3 mm^2. Secondly the method makes it possible to compare the absorption properties of materials whose scattering cross-section is far greater. Thus samples of graphite, whose absorption properties are of great importance to the nuclear power industry, can be compared to 1% by using 33 kg samples even though the scattering cross-section is 1000 times the absorption cross-section. Gleep is therefore used to a great extent for the routine testing of both uranium and graphite.

In more basic neutron physics it has been used for many thermal neutron cross-section measurements, but perhaps the most useful application has been in observing the changes in nuclear properties of uranium bars with irradiation. This is an important field of study, since the rate at which the net neutron yield from a uranium bar falls off with irradiation can have a marked effect on the cost of fuelling nuclear reactors.

Other uses of GLEEP which must be mentioned are as a source of neutrons for studies (*a*) of thermal neutron flux distributions in small sections of heterogeneous reactors and (*b*) of fast neutron effects on rats and mice.

BEPO

Reactor BEPO, too, is natural-uranium fuelled, graphite-moderated and air-cooled, but the cooling system provided is capable of removing 6000 kW of heat and so the maximum thermal neutron flux level reached is $1{\cdot}5 \times 10^{12}$ n cm^{-2} s^{-1}. As in GLEEP, the channels into which uranium is inserted are horizontal. There are 888 channels loaded, and the uranium is entirely of metal and is canned in aluminium with finned surfaces to increase the heat transfer surface, and prevent oxidation of the uranium. Charge and discharge of uranium is through holes in the end shields of the reactor, the shields consisting of 6 in. of iron followed by 6 ft. 6 in. of a barytes aggregate concrete.

Through the other three faces of the pile are a total of 60 experimental holes into which samples can be inserted or from which beams of neutrons can be allowed to emerge. At one side of BEPO a 10 × 10 ft column of graphite penetrates the concrete shield and is shielded at its outer end by a 1 ft door of cadmium-faced lead. A similar "thermal column" 6 × 6 ft emerges through the top shield. The main graphite block is a 26 ft cube with the loaded region forming a 20 ft diameter, 20 ft long cylinder in its centre. The cooling air is drawn in through a filter along underground ducts, up into a header space between the graphite and the shield over the uranium and out through another duct in which a filter and heat exchanger are installed, and thence to atmosphere through the exhausters and a stack 200 ft high.

Control of the reactor is exercised by slow insertion and removal of four horizontal steel tubes packed with boron carbide. As an emergency device ten similar tubes can be injected from the pile top to its centre is less than a second. The neutron flux level, and hence heat output, is observed with boron-coated ionization chambers, whose current output is amplified and recorded in the control room. Other measurements made continuously are temperatures of a number of uranium cartridges, temperature of the graphite at a number of places and of the air at inlet and outlet. The outgoing air is also checked for bulk activity and sampled for fission product activity check.

BEPO has been used as a reactor study in itself (temperature effects on reactivity, ^{135}Xe poisoning, γ-ray and neutron distribution studies, shielding studies) as a source of neutron beams and for numerous "in pile" irradiations. As an example of the use of a neutron beam the mechanical velocity selector or "chopper" may be mentioned. The first of these used on BEPO had a 2 m flight path and was used for such measurements as the total cross-section of ^{239}Pu, ^{235}U, ^{240}Pu as a function of neutron energy between 10^{-3} eV and 0·25 eV. A second higher speed chopper with a 33 m flight path is now in use on BEPO and using this it has been possible to improve resolution and extend the range of energy to between 10^{-2} eV and 100 keV.

Among the in-pile irradiations are the many different radio-

Table 1. *Summary of current uses of BEPO*

General description	*Number of holes*	*Typical use*
Isotope irradiations	14	^{32}P production
Fast chopper	1	Total neutron cross-section for ^{240}Pu in the eV region
Slow chopper	1	Neutron interaction with liquid hydrogen
Crystal spectrometers	3	Variation of σ fission/σ absorption with neutron energy
High-speed "rabbit"	1	Irradiation of fissile materials for out-of-pile delayed neutron energy and abundance studies
Neutron beams	6	Neutron slowing down studies
Oscillators	2	Measurement of η for fissile materials in "hot" neutron spectra
Radiation damage studies in a controlled environment	4	Creep of uranium under irradiation
Reactor "loop" experiments	3	Graphite–carbon dioxide reaction under irradiation

isotopes produced and the observations of the effect of neutron irradiation on the physical properties of materials such as graphite. Of a somewhat different character is the first major loop experiment conducted at Harwell, in which a simulated channel of a Calder Hall reactor was inserted in BEPO inside a steel tube. Carbon dioxide at the appropriate pressure is circulated through this loop and the reaction between graphite and carbon dioxide is studied under radiation conditions.

Other uses of BEPO are briefly summarized in Table 1.

DIMPLE

Reactor DIMPLE is simply an aluminium tank surrounded by graphite which is connected by stainless steel piping to a pump and a closed stainless steel tank. Heavy water can be raised by the pump from the stainless steel tank to the aluminium tank. A number of cadmium rods are suspended from the top of the aluminium tank on cables so that they can be dropped into the tank as required. Any desired arrangement of fuel can be placed in the aluminium tank so that DIMPLE is a very flexible system for the study of any desired heavy-water moderated reactor. The neutron flux level permitted is a maximum of 10^8 n cm^{-2} s^{-1} and is observed by boron chambers in the graphite reflector. The core features of interest are: critical size, neutron flux distribution, neutron spectrum and neutron fates.

All of these are studied in DIMPLE, the emphasis varying with the core under study. So far one natural uranium core, two slightly enriched cores and a series of highly enriched cores have been studied. In addition, one special core loading has been used to produce a pure thermal neutron spectrum in which to carry out some oscillator studies.

DIDO AND PLUTO

These two reactors are under construction at the moment. They are to be heavy-water cooled and moderated reactors using fuel containing a high proportion of ^{235}U. This uranium (2·5 kg of ^{235}U per charge) will be alloyed with aluminium and rolled into plates which are brazed together to form a fuel box of nine or ten plates. The active length of these boxes is 2 ft and twenty-five such boxes are supported at a 6 in. spacing in the centre of a 6 ft diameter 6 ft high tank. The top and bottom of the fuel plates are brazed into aluminium tubes so that heavy water can be pumped up through them to spill into the large tank before being pumped round again. This heavy water is cooled in heat-exchangers by a separate water circuit which disposes of its heat to atmosphere through cooling towers. At full power, 10 MW of heat will be removed and a maximum flux of 10^{14} n cm^{-2} s^{-1} will be possible.

Table 2. *Summary of proposed initial use of DIDO*

General description	*Number of holes*	*Typical use*
Radio-isotope production	12	High specific activity ^{60}Co production
Fast chopper	1	Neutron cross-section data up to 1 MeV
Slow chopper	1	Neutron interaction with liquid helium
Crystal spectrometers	2	Neutron diffraction in polycrystalline materials
Neutron beams	4	Neutron inelastic scattering time of flight spectrometer
Radiation damage studies	15	Damage during burn-up of fissile materials
Radiation chemistry	6	Uranyl sulphate solution decomposition and corrosion effects at temperature, pressure while under irradiation
Reactor "loop" experiments	3	Corrosion by, and decomposition of, high-temperature water in containers of various grades of steel

The differences between DIDO and PLUTO are in the experimental facilities provided by them. Broadly, DIDO is provided to satisfy the need for high intensity neutron beams, to produce high specific activity radio-isotopes and to enable small-scale, high-flux irradiations to take place. PLUTO, which will be ready one year after DIDO, is provided with a smaller number of large holes so that about six large-scale high-flux loop tests can be run simultaneously. During the interval between DIDO operating at high flux, and PLUTO becoming available, at least one large-scale loop will be operated in DIDO.

A summary of the initial programme for DIDO is presented in Table 2 as an indication of the range of requirements it has to satisfy.

LIDO

This reactor gets its name from the fact that it operates in a concrete water tank with internal dimensions 8 × 23 × 28 ft. The fuel elements are of the same general pattern as for DIDO, though since the moderator is natural water the proportion by volume of the moderator must be much less. The spacing of plates in the boxes and the spacing between boxes in the reactor core are therefore less than in DIDO. The core size will vary somewhat with the reflector and core arrangement, but will generally be in the range of 3 to 4 kg of ^{235}U. The core is suspended on a gantry which can move the length of the water tank from a trolley which can traverse the gantry. In this way the reactor core can be positioned against any of three aluminium panels 8 × 8 ft in the concrete wall of the tank, or against any of three holes each 12 in. in diameter. The reactor can therefore be used by erecting an experiment against a panel or hole in the concrete wall or by "canning" the experiment in a water-tight container and lowering it into the water.

At 100 kW heat output, the convective flow of water through the fuel element boxes will keep them adequately cooled. The maximum thermal neutron flux under these conditions will occur outside the core and be about 10^{12} n cm^{-2} s^{-1}.

It is proposed to use this reactor primarily for the development of shields for mobile reactors. This work is necessary since it is not yet possible to predict the amount of material required to produce fast neutron attenuation factors of the order of 10^8. Since γ-rays must also be removed, reactor shields are necessarily mixtures of light and heavy elements, and in trying to achieve a minimum weight or thickness of material, complex arrays of the two may be necessary.

Although such shielding studies will occupy much of the reactor time, its use as a general radiation source will be important. Experiments of this nature already planned include the construction, at one of the large panels, of sections of proposed reactor designs for which a direct measurement of the neutron spectrum is required. The core section under

study will be sub-critical but "driven" by the LIDO core and examined with a neutron spectrometer.

ZEPHYR AND ZEUS

These reactors are both zero-energy fast reactors. ZEPHYR is fuelled with plutonium, while ZEUS is fuelled with ^{235}U. They have been described by Shepherd.[2] Both were built to enable reactor studies to be made. The type of information required which cannot be obtained with sufficient accuracy in any other way includes (*a*) the neutron energy spectrum, (*b*) the breeding ratio, (*c*) the effect of various materials of construction on fast reactors, (*d*) the kinetics of fast reactor operation.

The results of studies in these categories are not generally fundamental nuclear properties of elements concerned. They are more usually "integral" quantities involving reaction rates in complex spectra, perhaps with several processes contributing. Such information is very valuable if the reactor under study (say ZEUS) closely resembles a high-power reactor (in this case the Dounreay fast reactor) which it is proposed to build.

SUMMARY OF THE U.K. REACTOR USES

The reactors described fall into four main categories according to their use, some of them combining two or three of these uses. The main categories are:

1. As a reactor core study (BEPO, DIMPLE, ZEPHYR, ZEUS).
2. As a measuring instrument (GLEEP, BEPO, DIMPLE).
3. As a neutron source for beams for physical studies, radio-isotope production, radiation damage tests, radiation chemistry (GLEEP, BEPO, DIDO, LIDO).
4. As a full-scale high-flux loop-testing machine for the nuclear power programme (BEPO, DIDO, PLUTO 1 and 2).

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DISCUSSION ON SESSION 8

Prof. J. Diamond: One of the most striking features of Dr. Lewis's and Mr. Fenning's papers is the evidence of the value of the first research reactors built in Canada and in the United Kingdom: NRX which began work nine years ago and BEPO eight years ago yesterday. Both are still in active use and are likely to be for a long time; both have yielded information of value in the international as well as the national scale and both have the immense advantage, in research reactors, of flexibility.

As Dr. Lewis says, NRX was for several years the most powerful irradiation tool in the world. Great credit is due to its designers who had to work at a time when little was known about reactor behaviour. BEPO is the only reactor which still appears in all four of Fenning's categories of uses. It is also noteworthy that each reactor seems to have founded the line of nuclear power development which has been adopted in each country: NPD and Calder Hall are direct descendents of NRX and BEPO.

In this country we see a growth of two distinct groups of research reactors—those in which the prime function is the production of a high flux for irradiation work (DIDO, PLUTO) and those specialist reactors such as DIMPLE, ZEPHYR and ZEUS which are themselves experiments in reactor technology.

As knowledge and programs have grown, the need has arisen for higher neutron fluxes. In both countries this is being met by heavy water reactors. It is not at all clear what further developments are likely to take place in this group. There is always a desire for more space and more flux, but it becomes very expensive to exceed the 10^{14} n cm^{-2} s^{-1} of DIDO and PLUTO. We have heard that NRU will yield 3×10^{14} n cm^{-2} s^{-1} maximum flux, but here the cost is offset to some extent by the plutonium and other isotopes which will be made. Until there is some likelihood of power reactors operating at such a flux the purely technological value of, say, 10^{15} n cm^{-2} s^{-1}, especially if it is achieved at the sacrifice of space, may be in some doubt, but from the point of view of basic research, which inevitably finds its way into technology, there is reason to attempt its achievement, despite the enormous cost which would be involved—at least, with solid fuel elements.

One wonders whether we must wait for liquid fuel reactors before it would be wise to make the attempt.

It is always difficult to distinguish between "basic" and "technological" research and it is interesting to note from Table 2 of Mr. Fenning's paper that some 80% of the holes in DIDO will be devoted to what are primarily production and technological studies.

To return to irradiation work of the kind which will be carried out in PLUTO, it would be interesting to know what the authors feel about the value of higher fluxes than 10^{14} n cm^{-2} s^{-1} in irradiation work now and in the near future, observing, as was mentioned by Grout at the Inaugural Meeting of the British Nuclear Energy Conference* this year, that in the search for long-term effects it is frequently undesirable to shorten irradiation times by exposing specimens to higher fluxes than those for which they are intended in service.

In addition to the uses of the research reactors described in the papers, mention should be made of the part they have played and will continue to play in the training of staff. BEPO and NRX have been of particular value in this respect. It has been said that few of the U.K. Atomic Energy Authority staff have ever operated a reactor, nor are they likely to do so. This must be as much due to shortage of reactors as to anything else and I feel certain that with the increased opportunities for work *on and around* reactors more of the engineering and scientific staff of the Authority and industrial firms will be able to acquire firsthand knowledge of reactor operation and a "feel" for the work to the great advantage of everyone concerned with these developments.

Need I say that those whose business it is to educate and train would be glad of such opportunities?

Mr. J. J. McEnhill: A feature common to both Dr. Lewis's and Mr. Fenning's papers is the intimate connection which research reactors in both the U.K. and Canada have had with the provision of information on reactor development for the power production programme of both countries. Some purists would protest that some of these reactors are not strictly in the research reactor category, but nevertheless all

* GROUT, H. J. *J. Brit. Nuclear Energy Conf.*, **1**, p. 35 (1956).

of them have embodied research facilities which have enabled both countries to stand well in the forefront of all forms of nuclear research both fundamental and applied.

However, all of these facilities are in Government establishments. The time has now arrived when thoughts are turning to a more diverse location and there are signs that this is now happening in the U.K.; for example, the plans for the swimming pool reactor MERLIN at Research Laboratories of Associated Electrical Industries Ltd., and some universities have also been considering the problem seriously. Because of high capital and running costs with complex reactors, such facilities would have to be simpler, inherently safe, and the investment of excess reactivity quite small. Simple zero energy thermal neutron systems are useful tools in reactor physics research, e.g. when used in conjunction with pulsed neutron sources and as there is scope for such research these would provide a useful university facility where their application in the teaching of the elements of reactor control to engineers would also be invaluable. Perhaps Mr. Fenning would care to comment on this and also on the problems associated with the installation and operation of medium power reactor systems, e.g. of swimming pool or water boiler types in universities.

Mr. Fenning mentioned the applications of LIDO to the study of proposed reactor design sections, especially for examination of their neutron spectrum. Does he imply that a subcritical reactor section of relatively high neutron multiplication will be used with a large area thermal neutron source of fairly low intensity from LIDO to drive it? If so, will not the spectrum be contaminated by the source neutrons, and how is the information which is obtained to be applied to core design?

Dr. Laurence's description of the control of the NPD reactor by variations of the heavy water level is very interesting. He suggests that normal absorber rod control is unsatisfactory for measurements which depend on a steady flux distribution within the core. DIDO uses "signal arm" controls so that presumably this reactor will suffer from this disadvantage. Mr. Fenning might perhaps care to comment on this?

Dr. A. K. Krasin: In the Soviet Union there are reactors equipped for physical and materials-testing research. Information about these reactors has been given in the Geneva Conference reports and in articles in the journal *Atomnaya Energiya*.

There are also reactors of the so-called zero-energy type. The reactor with a moderator of metallic beryllium is of this type. The moderator of this reactor is disposed in the form of a cylinder. The cross-section is given in Fig. 1.

In the beryllium reactor uranium containing 10% of the isotope ^{235}U is used. The enriched uranium was stacked in the reactor in tubular elements. A cross-section of an element and the scheme used for the cells are shown in Fig. 2.

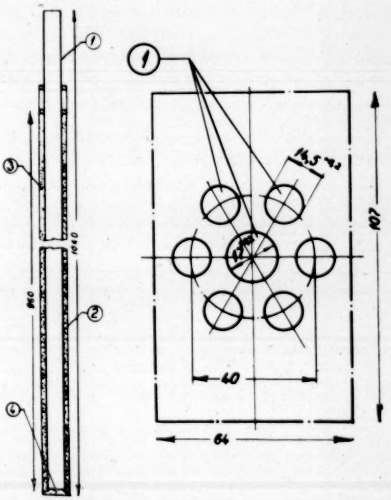

Fig. 2. Cross-section of element of reactor shown in Fig. 1

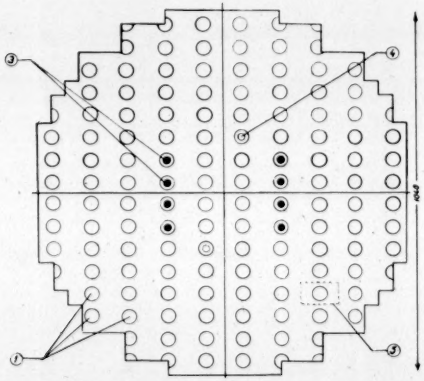

Fig. 1. Cross-section of zero-energy type USSR reactor

Some results of measurements are given in the table which contains data for critical masses of some variants of the reactor.

The theoretical values of the critical masses were obtained by the multi-group method.

Type of reactor	*Critical mass of ^{235}U (kg)*	
	exp.	*theor.*
Dry reactor, without reflector	11·7	12 ± 1·6
Dry reactor, with reflector at sides	6·7	7 ± 1·4
Dry annular reactor	12·7	—
Reactor with water inside uranium elements	3·4	—

More detailed data will be printed in the journal *Atomnaya Energiya*.

Mr. I. J. W. Bisset: The foundations of a 10000 kW heavy water moderated research reactor almost identical with DIDO at Harwell has been completed on part of an 80 acre site at Lucas Heights near Sydney, New South Wales, Australia. It is hoped to complete construction of the reactor by mid-1957 and have it at full power early in 1958. I wish to express our appreciation of the considerable help that

Australia has received from the U.K.A.E.A. with the reactor project. Hot chemistry metallurgy and other research laboratories are being simultaneously constructed under a £A5½ million project approved by the Cabinet of the Commonwealth Government in September 1954. Some thirty Australian scientific officers under their Chief Scientist Mr. C. N. Watson-Munro are in the meantime working at Harwell.

Mr. J. Bussac: The research reactors in operation in France are the low power ZOE reactor, and the 10^{13} n cm^{-2} s^{-1} flux P2 reactor. In the near future we shall have a zero power research reactor, named AQUILON, to give information for heavy water lattices, and towards the end of 1957 we hope to have a high flux (10^{14} n cm^{-2} s^{-1}) heavy water slightly enriched uranium reactor, which will be similar to DIDO.

I wish to point out the difficulty that may be caused by radiation damage on the pressure vessel of reactors. For the same power, the core of a heavy water reactor is bigger than that of a light water one. What is Dr. Laurence's opinion on that problem? Will the radiation damage of pressure vessels be a future limitation of heavy water power reactors by making a thick reflector very expensive and requiring a very large pressure vessel? Can Dr. Laurence give the thickness of the reflector designed for the NPD reactor, and does he think that the 1000 MW reactor described at the Geneva Conference would be safe from the point of view of Wigner effects on the vessel?

Dr. L. Kowarski: The French version of DIDO is considerably less enriched, and therefore has a higher power (15–20 MW). In this sense it can be said to be intermediate between NRU (natural uranium) and DIDO or CP5 (fully enriched), and it will be interesting to see to what extent it combines the advantages of both types. The fuel elements are tubular and the flow of coolant is quite different from that of the DIDO type of reactor.

Dr. J. R. Dietrich: Since the question of low-power research reactors has been brought up in the previous discussion I would like to remark that the water-boiler, swimming-pool, and CP5 (heavy water-cooled enriched uranium) types are in use or under construction or design by universities in the United States, notably by North Carolina State University, Pennsylvania State University, University of Michigan, and Massachusetts Institute of Technology. Of these reactor types, perhaps only the water-boiler is really a low-power type. The Argonne National Laboratory, in connexion with its operation of the International Reactor School, is currently designing and building a low-power reactor (the Argonaut Reactor) which will be suitable for reactivity and control measurements, and for use as a neutron source for sub-critical assemblies. It is believed that such a reactor meets a need which cannot conveniently be met by the higher-power reactor types. Much of the design effort is being directed toward minimizing the cost of the reactor.

Dr. A. Salmon: I should like to ask Dr. Laurence if the reactor analogue briefly mentioned in Dr. Lewis's paper could be described; if there had been any evidence of Wigner growth or stored energy in the NRX thermal columns, and, in the light of their experimental experience, was there a real requirement for two thermal columns in NRX. Could Dr. Dietrich say if there was any evidence on the transient behaviour of water-cooled and moderated reactors when there was a considerable depth, say 20 ft, of water above the core? This was an important case as many "pool" reactors were now being built which would have such a depth of water; experimental evidence on their transient behaviour would be of great value.

Dr. J. R. Dietrich: I believe that the effect of depth of submersion has been investigated, over the relatively small range of zero to about four feet, as part of the SPERT programme which is being carried out by the Phillips Petroleum Company at the National Reactor Testing Station in Idaho, and that greater depths, which require modification of the existing equipment, will eventually be investigated. I believe that no important effect was observed over the range tested. Theoretically, of course, one would expect an increase in depth of submersion to cause some increase in the quantity of energy released during a power excursion terminated by steam formation in the reactor. This increase would be the result of an increase in saturation temperature at the reactor core location, partly because of the static pressure of the water above the core and partly because of the dynamic pressure generated in accelerating the water as steam is formed in the core.

Mr. S. A. Skvortsov: In the Soviet Union great attention is paid to the educational matters associated with nuclear energy projects. In this connexion some reactor designs have been developed for technical colleges and universities for preparing and training physicists. Their reactors with light and heavy water moderators will have heat capacities of 100–5000 kW. They will be supplied with holes to release neutron beams and with openings for charging samples to be tested. Reactors of this kind will be useful not only for educational purposes but for the development of scientific investigations. Some of these reactors are about to be constructed.

Dr. G. C. Laurence: Replying to Professor Diamond's question on Dr. Lewis's paper regarding the adequacy of operating flow area surrounding the NRU reactor, the main operating hall is 175 ft long and 160 ft wide. I can remember the planning and construction of a large number of research laboratories in the past; in every case the space provided seemed to be more than adequate during planning but within a few years was found to be insufficient. I have no doubt that this experience will be repeated with the NRU reactor.

Replying to Dr. Salmon regarding difficulties on the storage of energy in the graphite of the thermal columns of NRX, I have no evidence that this was discovered when the reactor was taken apart for repairs following the accident of December 1952.

Commenting on the usefulness of the thermal columns of NRX for research purposes, I wish to say that there has not been as much demand for their use as had been expected when the reactor was designed. However, they are particularly suitable for certain types of experiment to justify the inclusion of a large thermal column in the construction of NRU.

An analogue computer has been found extremely useful at Chalk River for rapid calculations of flux distribution in reactor lattices of non-uniform composition. The analogue was used to simulate 2-group analysis of the flux distribution in a one dimensional lattice. Thermal flux and fast neutron flux were represented by potentials at 20 lattice positions. After six months' experience with this instrument it is regarded as a very useful tool and as finding application in other problems such as heat transfer.

Replying to Mr. Bussac I am far from satisfied with the extent of knowledge of the effects of radiation damage in the steel of heavy pressure shells used to enclose reactors. However, it is felt that the shell of the NPD reactor is reasonably safe in this respect. It is not too difficult to fabricate pressure shells up to 11 or 12 ft in diameter and with a wall 6 in. thick. The cost of manufacture will increase rapidly as these dimensions are exceeded. As an alternative to the use of a

pressure shell, the coolant might be confined in pressure tubing surrounding a few elements. The objection to this is the wastage of neutrons by capture in these thick walled-tubes.

In NPD reactor the thickness of heavy water between the fuel rods of the core and steel radiation shield just inside the vertical wall of the reactor vessel is about 20 cm. This gap is dictated by reasons of mechanical design, but of course acts as a thin reflector.

AUTHOR'S REPLY

Mr. F. W. Fenning. In answer to Prof. Diamond's question about the value of higher fluxes, I do not think that fluxes in excess of 10^{14} n cm^{-2} s^{-1} are vital to present technological studies. However, it must be emphasized that with DIDO, although the flux is 10^{14} n cm^{-2} s^{-1} in the fuel, it will in general be less in a large scale test sample. For in creating the correct environment for the test, it is usually necessary to introduce a large hole in the moderator and a steel pressure tube may be required with the effect of reducing flux at the test sample. Therefore some increase in flux in a DIDO type core might be desirable in those cases introducing the worst depressions. In addition, we should note that the comments of Mr. Grout referred to by Prof. Diamond do not apply to many research items which become easier or quicker to study as fluxes are increased.

Mr. McEnhill invited me to comment on the use of zero energy and higher power reactors in university teaching. I feel that the physicist has much to learn from zero energy assemblies, but the engineer much more from higher power reactors. But, whereas a zero energy assembly could be within the scale of expenditure usually undertaken by a university department, the higher power reactor will inevitably involve large capital expenditure, and high operating cost if it is to be safely operated. For reactor control studies, the electronic analogue has much to recommend it in university teaching.

With regard to the use of LIDO to "drive" a sub-critical section of a lattice for spectrum measurement, it is of course essential to choose the dimensions of such a lattice to ensure the elimination of primary source neutrons.

Mr. McEnhill's remarks about the effect of DIDO control arms are very relevant. Large flux distortions will certainly be caused, and we regard it as one of the most important operational requirements to learn to operate the reactor so that the control arms spend most of their time towards the outside of the core.

Exhibits and films

EXHIBITS FROM HARWELL

The United Kingdom Atomic Energy Authority's Research Establishment at Harwell provided the following exhibits which were on view throughout the Conference.

Čerenkov radiation and light flashes from the night sky. This exhibit showed the light receiver and recording apparatus used by Drs. Galbraith and Jelley at the Pic du Midi Observatory in France, for their work on light flashes from the night sky caused by Čerenkov radiation in the atmosphere associated with cosmic-ray air showers. A tableau also described Čerenkov radiation, its practical applications in high-energy physics, its occurrence in water-moderated reactors and the special case of the light produced in the atmosphere.

(An account by Dr. Jelley of this radiation and its application was published in the *British Journal of Applied Physics* for July 1955.)

A compact high-voltage d.c. generator. A compact unit was displayed which provides a source of high voltage for applications where currents up to 0·5 mA may be required. The unit was originally developed as a compact high-voltage supply for the focusing electrodes associated with the ion source of the proton linear accelerator being constructed at Harwell.

The reactors LIDO and DIDO. Models of new reactors being built at Harwell were shown. LIDO is a thermal reactor (100 kW) with solid enriched uranium plate type fuel elements and has a flux of 10^{12} n cm^{-2} s^{-1}. It will be moderated and cooled by circulated and filtered demineralized, light water. It has been designed for the investigations of shielding problems.

DIDO is a heavy-water cooled and moderated reactor and will use highly enriched uranium, alloyed with aluminium, as fuel. With its high intensity neutron flux of 10^{14} n cm^{-2} s^{-1} at a designed heat output of 10000 kW, the reactor's main purpose will be the irradiation of materials to be used in future reactors.

Metallurgical exhibit. Some of the fabrication techniques involved in reactor construction were illustrated. The materials used include uranium, thorium, zirconium, beryllium and some refractory compounds of these metals. Results of some neutron diffraction studies were displayed and some examples shown of the effect of irradiation upon gemstones. A simplified model of the disordering of solids by atomic radiation was also on show.

FILMS

The coloured film showing the construction of the first United Kingdom nuclear power station at Calder Hall was shown.

Through the courtesy of Dr. J. R. Dietrich a selection of films about experiments with the boiling water reactors of the Argonne National Laboratory, Illinois, was shown. Copies of one of the films, with commentary, are available from the U.S. Atomic Energy Commission, Washington 25, D.C., U.S.A.

BOOKS AND PERIODICALS

Examples of the Institute's publications were shown, and there was a display of other books and periodicals cognate to the subject of the Conference.